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TITLE: ORGANIC ELECTROLUMINESCENCE ELEMENT MATERIAL  
AND ORGANIC ELECTROLUMINESCENCE ELEMENT OBTAINED USING THE  
SAME

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ABSTRACT:

PROBLEM TO BE SOLVED: To obtain an organic electroluminescence element having high light- emitting efficiency and excellent in stability when repeatedly used.

SOLUTION: The organic electroluminescence element material is represented by formula [1], wherein at least seven of R1-R12 are each an aryl group and all of R1 through R4 are not hydrogen atoms simultaneously.

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CLAIMS DETAILED DESCRIPTION TECHNICAL FIELD PRIOR ART EFFECT OF THE INVENTION  
TECHNICAL PROBLEM MEANS EXAMPLE

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[Translation done.]

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3. In the drawings, any words are not translated.

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**DETAILED DESCRIPTION**

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the organic electroluminescence (EL) component used for the flat-surface light source or a display.

[0002]

[Description of the Prior Art] Promising \*\* of the application as a large area full color display device with a cheap solid-state luminescence mold is carried out, and, as for the EL element which used the organic substance, many development is performed. Generally the EL element consists of counterelectrodes of the pair the luminous layer and this whose layer were pinched.

[0003] When electric field are impressed between two electrodes, an electron is poured in from a cathode side and, as for luminescence, an electron hole is poured in from an anode plate side. Furthermore, in case this electron recombines with an electron hole in a luminous layer and an energy level returns from a conduction band to a valence band, it is the phenomenon which emits energy as a light.

[0004] Compared with the inorganic EL element, the conventional organic EL device had high driver voltage, and luminescence brightness and its luminous efficiency were also low. Moreover, property degradation is also remarkable and it had not resulted in utilization.

[0005] In recent years, the organic EL device which carried out the laminating of the thin film containing an organic compound with the high fluorescence quantum efficiency which emits light by the low battery not more than 10V is reported, and the interest is attracted (refer to applied FIJIKUSU Letters, 51 volumes, 913 pages, and 1987).

[0006] By this approach, the fluorescent substance layer and the amine system compound were used for the hole-injection layer for the metal chelate complex, green luminescence of high brightness has been obtained, 100 cd/m<sup>2</sup> and the maximum luminous efficiency attain 1.5 lm/W with the direct current voltage of 6-7V, and brightness has the engine performance near a practical use field. However, although luminescence reinforcement is improved for the organic EL device to current by the improvement of a configuration, it does not have still sufficient luminescence brightness. Moreover, it has the big problem of being inferior to the stability at the time of repeat use.

[0007]

[Problem(s) to be Solved by the Invention] The purpose of this invention has high luminous efficiency, and is in offer of the organic EL device which was excellent in the stability in the time of repeat use. as a result of this invention persons' inquiring wholeheartedly, the luminous efficiency of the organic EL device which boiled further the organic EL device ingredient of the compound shown by the general formula [1], and used it at least was high, and it found out that the stability in the time of repeat use was also excellent, and resulted in this invention.

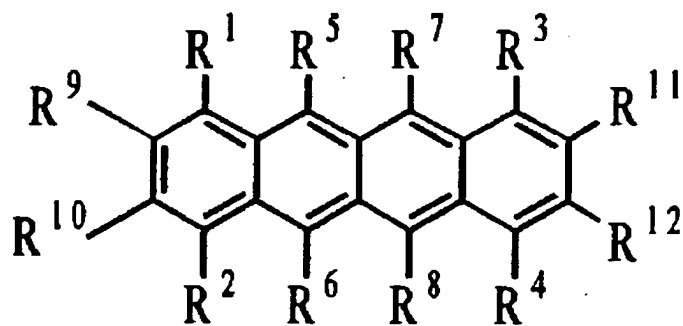
[0008]

[Means for Solving the Problem] That is, this invention relates to the organic electroluminescent element ingredient shown by the following general formula [1].

[0009] General formula [1]

[0010]

[Formula 2]



[0011] Inside of [ , however a formula R1-R12 express the organic residue which is not permuted [ the permutation chosen from the group which consists of a hydrogen atom, a halogen atom or an alkyl group, an aryl group an alkoxy group, an aryloxy group, an alkylthio group, an aryl thio radical, an amino group, and heterocycle residue independently, respectively, or ], it may join together by the substituent comrade who approached, and R1-R12 may form a new ring. Moreover, at least seven of R1-R12 pieces are the aryl group which is not permuted [ a permutation or ], and all of R1-R4 do not become a hydrogen atom at coincidence.] Moreover, this invention relates to the organic electroluminescent element which is the layer in which at least one layer contains the above-mentioned organic electroluminescent element ingredient in the organic electroluminescent element in which organic compound thin film much more at least which contains a luminous layer in inter-electrode [ of a pair ] was formed.

[0012] Moreover, this invention relates to the organic electroluminescent element which is the layer in which a luminous layer contains the above-mentioned organic electroluminescent element ingredient in the organic electroluminescent element in which the organic compound thin film of two or more layers which contains a luminous layer in inter-electrode [ of a pair ] was formed.

[0013] Moreover, this invention relates to the organic electroluminescent element which is the layer in which at least one layer of the hole-injection band between a luminous layer and an anode plate contains the above-mentioned organic electroluminescent element ingredient in the organic electroluminescent element in which the organic compound thin film of two or more layers which contains a luminous layer in inter-electrode [ of a pair ] was formed.

[0014]

[Embodiment of the Invention] R1-R12 in the compound expressed with the general formula [1] of this invention express independently the organic residue which is not permuted [ a hydrogen atom, a halogen atom, the permutation shown in the following, or ], respectively.

[0015] With the organic residue of this invention, an alkyl group, an aryl group, an alkoxy group, an aryloxy group, an alkylthio group, an aryl thio radical, the amino group, and heterocycle residue are mentioned.

[0016] As an aryl group in this invention, a phenyl group, a biphenyl radical, a terphenyl radical, A quarter phenyl group, o-, m- and p-tolyl group, a xylyl group, o-, m- and p-KUMENIRU radical, a mesityl radical, a pentalenyl radical, An indenyl group, a naphthyl group, a BINAFTURE nil radical, a TANAFUTARE nil radical, A quarter North America Free Trade Agreement RENIRU radical, an azulenyl radical, a hepta-RENIRU radical, a BIFE elm nil radical, An in DASENIRU radical, a fluoran thenyl radical, an ASENAFUCHIRENIRU radical, the ASEAN TORIRENIRU radical, A phenalenyl radical, a fluorenyl group, an anthryl radical, a BIANTORASENIRU radical, A TAANTORASENIRU radical, a quarter anthracenyl group, an anthra quinolyl radical, A phenan tolyl group, a TORIFE elm nil radical, a pyrenyl radical, a chestnut SENIRU radical, The North America Free Trade Agreement SENIRU radical, a play adenyl radical, a PISENIRU radical, a peri RENIRU radical, a PENTA phenyl group, There are a pen TASENIRU radical, a TETORAFE elm nil radical, a hexa phenyl group, a hexa SENIRU radical, a ruby SENIRU radical, a koro NENIRU radical, a TORINAFUCHIRENIRU radical, a hepta-phenyl group, a hepta-SENIRU radical, a pyran TORENIRU radical, an OBARENIRU radical, etc.

[0017] As heterocycle residue in this invention, moreover, a thienyl group, a benzo [b] thienyl group, A [2 and 3-naphth b] thienyl group, a CHIANTORENIRU radical, a furil radical, a pyranil radical, An iso benzofuranyl radical, a clo MENIRU radical, a KISAN thenyl radical, a FENOKISA tsi nil radical, A 2H-pyrrolyl radical, a pyrrolyl radical, an imidazolyl radical, a pyrazolyl radical, a pyridyl radical, A pyrazinyl radical, a pyrimidinyl group, a pilus DAJINIRU radical, an in DORIJINIRU radical, An iso indolyl radical, a 3H-indolyl radical, an indolyl radical, a 1H-indazolyl group, The Puri Nils radical, a 4H-kino RIJINIRU radical, an iso quinolyl

radical, a quinolyl radical, A phthalazinyl radical, a naphthyridinyl group, a kino KISANIRIRU radical, a chinae-cortex ZORINIRU radical, A SHINNORINIRU radical, a PUTERIJINIRU radical, a 4aH-carbazolyl radical, a carbazolyl radical, beta-cull BORINIRU radical, a phenanthroliziny radical, an acridinyl radical, a peri midge nil radical, A phenan trolley nil radical, a FENAJINIRU radical, a FENARUSAJINIRU radical, an iso thiazolyl radical, A phenothiazinyl group, an isoxazolyl group, a furazanyl group, a phenoxazinyl radical, An iso chromanyl radical, a chromanyl radical, a pyrrolidinyl radical, a pylori nil radical, There are an imidazolidinyl radical, an imidazo RINIRU radical, a PIRAZORIJINIRU radical, a PIRAZORINIRU radical, a piperidyl radical, a piperazinyl radical, an in DORINIRU radical, an iso in DORINIRU radical, a quinuclidinyl radical, a mol HORINIRU radical, etc.

[0018] Moreover, as an alkyl group in this invention, there are a methyl group, an ethyl group, n- and an iso-propyl group, n-, iso-, sec- and tert-butyl, n-, iso-, neo- and a tert-pentyl radical, n-, iso- and a neo-hexyl group, benzyl, etc.

[0019] As an amino group in this invention, the 1st class amino group, a monoalkylamino radical, a mono-arylamino radical, the mono-heterocycle residue amino group, a dialkylamino radical, the diaryl amino group, the II heterocycle residue amino group, an alkyl arylamino radical, the alkyl heterocycle residue amino group, and the aryl heterocycle residue amino group are mentioned.

[0020] The organic residue of R1-R12 may be permuted by other substituents among the compound expressed with a general formula [1]. As the class of substituent There is an alkylamino radical which is not permuted [ the amino group which is not permuted / the alkylthio group which is not permuted / the alkoxy group which is not permuted / the alkyl group which is not permuted / a halogen atom, a cyano group, a nitro group, a permutation, or /, a permutation, or /, a permutation, or /, a permutation, or /, a permutation, or /, a permutation, or ].

[0021] In this invention, the compound expressed with a general formula [1] is compoundable by the following approaches, for example. In addition, the synthesis method of a compound expressed with a general formula [1] is not limited to these.

[0022] An iso benzofuran derivative is made to react with dibromo ethylene, and a dibromo naphthalene derivative is compounded. This dibromo naphthalene derivative is made to react with an iso benzofuran derivative, and the compound expressed with a general formula [1] is compounded.

[0023]

[Example] The heating ring current of the synthesis method [ of 1, 4, 5, 6 7, 10, 11, 12, and 13-OKUTA phenyl naphthacene ] 2, 3, 6, and 7-tetra-phenyl iso benzofurans 3g (7.1 mmol) and 1, 2-dibromo ethylene 2.6 g (14.2 mmol), and the p-toluenesulfonic-acid 50 mg was carried out in xylene 15 ml for 12 hours. Then, the solid-state which added methanol 15 ml and deposited was \*\*\*\*(ed), and 2 and 3-dibromo -1, 4, 5, and 8-tetra-phenyl naphthalene 2.8 g was obtained by recrystallizing in benzene-ethanol (67% of yield). Next, 2 and 3-dibromo -1, 4, 5, and 8-tetra-phenyl naphthaleneg [ 6.8 ] (11.5 mmol) and 2, 3, 6, and 7-tetra-phenyl iso benzofuran 4.7 g (11.1 mmol) is dissolved in tetrahydrofuran 100 ml, and at -78 degrees C, n-butyl lithium 0.73 g (11.4 mmol) is dropped, and it warms to a room temperature, and agitates for 12 hours. Water 100 ml is added after reaction termination, hexane 100 ml is added further, and the solid-state which cooled and deposited to 0 degree C is \*\*\*\*(ed). 5, 12-epoxy -1, 4, 5, 6, 7, 10, 11, and 12 OKUTA phenyl naphthacene 8.2 g were obtained by recrystallizing this deposit solid-state in benzene-ethanol (89% of yield). Next, 5, 12-epoxy -1, 4, 5, 6, 7, 10, 11, and 12 OKUTA phenyl naphthacene 8.5 g (10.2 mmol) are dissolved in chloroform 50 ml, dibromomethane solution 3.75 ml which contains aluminium-bromide 0.4 g (3.75 mmol) at -78 degrees C is dropped, and it warms to a room temperature. Sodium disulfite water-solution 25 ml is added 5% after reaction termination, an organic layer is separated, and it dries with magnesium sulfate. Evaporation of the organic layer was carried out and 1, 4, 5, 6, 7, 10, 11, 12, and 13-OKUTA phenyl naphthacene 6.6 g were obtained by recrystallizing the residue in benzene-ethanol (80% of yield).

[0024] Although the example of representation of the compound of this invention is concretely illustrated to Table 1, this invention is not limited to the following examples of representation.

[0025] Table 1 [Table 1]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
1	H								H	H	H	H
2									H	H	H	H
3					H				H	H	H	H
4												
5											H	H
6			H	H								
7		H	H	H							H	H
8		H	H	H						H	H	

[0026]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
9			H	H					H	H		
10			H	H							H	H
11									CH <sub>3</sub>	CH <sub>3</sub>	H	H
12									CH <sub>3</sub>	H	CH <sub>3</sub>	H
13									CH <sub>3</sub>	H	H	CH <sub>3</sub>
14									OCH <sub>3</sub>	OCH <sub>3</sub>	H	H
15									OCH <sub>3</sub>	H	OCH <sub>3</sub>	H
16									OCH <sub>3</sub>	H	H	OCH <sub>3</sub>

[0027]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
17									CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
18									CH <sub>3</sub>	H	H	H
19									OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>
20									OCH <sub>3</sub>	H	H	H
21									SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>
22									SCH <sub>3</sub>	H	H	H
23									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
24									SCH <sub>3</sub>	H	SCH <sub>3</sub>	H

[0028]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
25									SCH <sub>3</sub>	H	H	SCH <sub>3</sub>
26	CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>								
27	CH <sub>3</sub>	H	H	H								
28	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
29	CH <sub>3</sub>	H	CH <sub>3</sub>	H								
30	CH <sub>3</sub>	H	H	CH <sub>3</sub>								
31	OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>								
32	OCH <sub>3</sub>	H	H	H								

[0029]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
33	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H								
34	OCH <sub>3</sub>	H	OCH <sub>3</sub>	H								
35	OCH <sub>3</sub>	H	H	OCH <sub>3</sub>								
36	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>								
37	SCH <sub>3</sub>	H	H	H								
38	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
39	SCH <sub>3</sub>	H	SCH <sub>3</sub>	H								
40	SCH <sub>3</sub>	H	H	SCH <sub>3</sub>								

[0030]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
41												
42		H	H	H								
43			H	H								
44		H		H								
45		H	H									
46	SCH <sub>3</sub>	SCH <sub>3</sub>										
47	CH <sub>3</sub>	CH <sub>3</sub>										
48	OCH <sub>3</sub>	OCH <sub>3</sub>										

[0031]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
49												
50										H	H	H
51											H	H
52										H		H
53										H	H	
54									SCH <sub>3</sub>	SCH <sub>3</sub>		
55									CH <sub>3</sub>	CH <sub>3</sub>		
56									OCH <sub>3</sub>	OCH <sub>3</sub>		



[0032]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
57												
58										H	H	H
59											H	H
60										H		H
61										H	H	
62									SCH <sub>3</sub>	SCH <sub>3</sub>		
63									CH <sub>3</sub>	CH <sub>3</sub>		
64									OCH <sub>3</sub>	OCH <sub>3</sub>		

[0033]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
65												
66		H	H	H								
67			H	H								
68		H		H								
69		H	H									
70	SCH <sub>3</sub>	SCH <sub>3</sub>										
71	CH <sub>3</sub>	CH <sub>3</sub>										
72	OCH <sub>3</sub>	OCH <sub>3</sub>										

[0034]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
73												
74										H	H	H
75											H	H
76										H		H
77										H	H	
78									CH <sub>3</sub>	CH <sub>3</sub>		
79									OCH <sub>3</sub>	OCH <sub>3</sub>		
80									SCH <sub>3</sub>	SCH <sub>3</sub>		

[0035]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
81												
82		H	H	H								
83			H	H								
84		H		H								
85		H	H									
86	CH <sub>3</sub>	CH <sub>3</sub>										
87	OCH <sub>3</sub>	OCH <sub>3</sub>										
88	SCH <sub>3</sub>	SCH <sub>3</sub>										

[0036]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
89									H	H	H	H
90									H	H	H	H
91									H	H	H	H
92									H	H	H	H
93									H	H	H	H
94											H	H





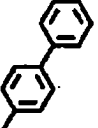
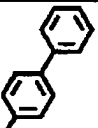
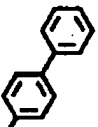
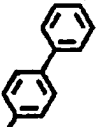
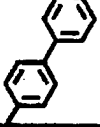
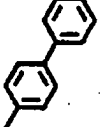
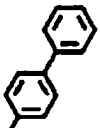
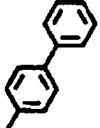








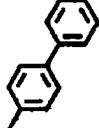
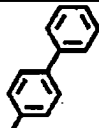
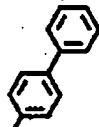
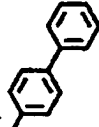
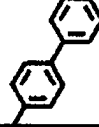
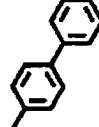
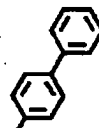
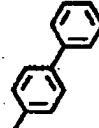








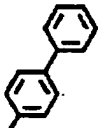
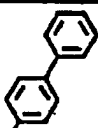
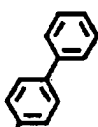
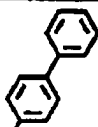
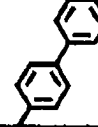
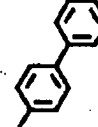
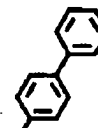
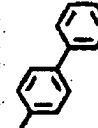




[0037]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
95			CH <sub>3</sub>	CH <sub>3</sub>					CH <sub>3</sub>	CH <sub>3</sub>		
96	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
97	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H								
98	CH <sub>3</sub>	CH <sub>3</sub>	H	H					H	H		
99	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
100			CH <sub>3</sub>	CH <sub>3</sub>								

[0038]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
101											H	H	H
102											H	H	H
103											CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
104											CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
105											CH <sub>3</sub>	H	H
106											CH <sub>3</sub>	H	H





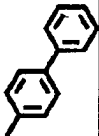
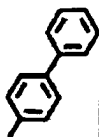


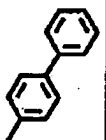
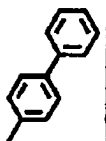










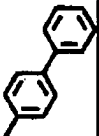
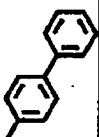


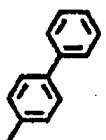
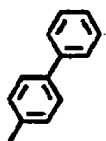





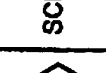




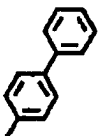
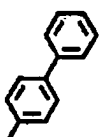


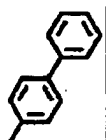
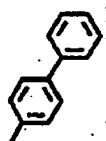

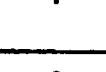



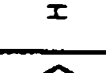
[0039]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
107									SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>
108									SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>
109									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
110									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
111									OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>
112									OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>

















































[0040]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
113									OCH <sub>3</sub>	OCH <sub>3</sub>	H	H
114									OCH <sub>3</sub>	OCH <sub>3</sub>	H	H
115									SCH <sub>3</sub>	SCH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
116									SCH <sub>3</sub>	SCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>
117									OCH <sub>3</sub>	OCH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
118									OCH <sub>3</sub>	OCH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>

[0041]





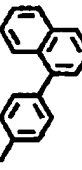
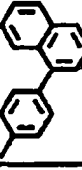




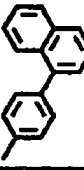
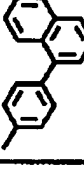









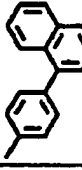




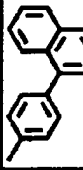
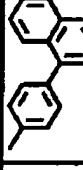








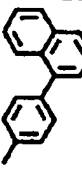





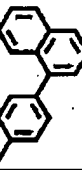
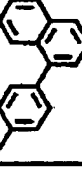




化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
119									OCH <sub>3</sub>	OCH <sub>3</sub>	H	H
120									CH <sub>3</sub>	CH <sub>3</sub>	H	H
121									H	H	CH <sub>3</sub>	CH <sub>3</sub>
122									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
123									OCH <sub>3</sub>	OCH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
124									H	H	CH <sub>3</sub>	CH <sub>3</sub>

[0042]





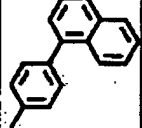
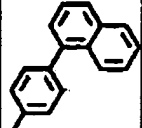
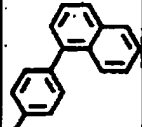
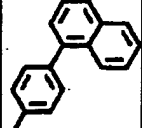
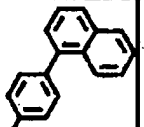
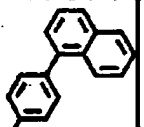
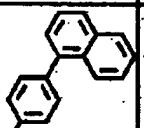
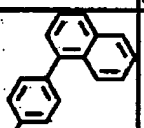








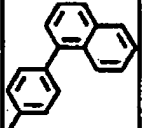
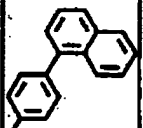
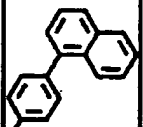
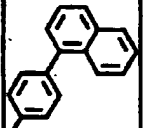
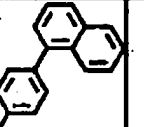
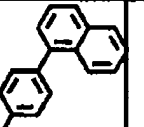
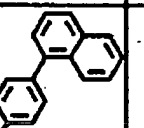
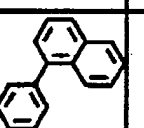








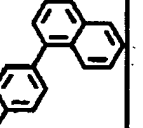
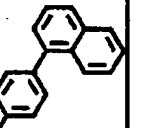
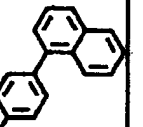
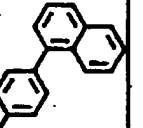
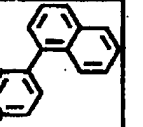
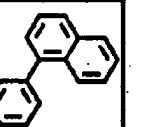
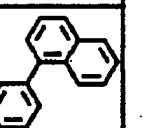
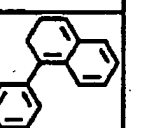




化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
125	C <sub>2</sub> H <sub>5</sub>	C <sub>2</sub> H <sub>5</sub>	H	H								
126	C <sub>2</sub> H <sub>5</sub>	C <sub>2</sub> H <sub>5</sub>	H	H								
127	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
128	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>								
129	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
130	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								

[0043]



















































化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
131	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
132	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
133	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
134	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>								
135	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
136	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								





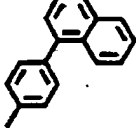
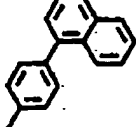
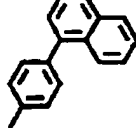
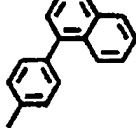
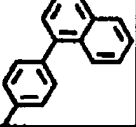
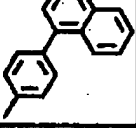
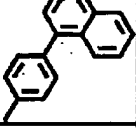
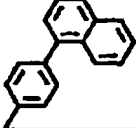








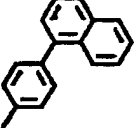
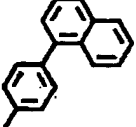
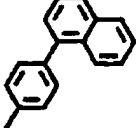
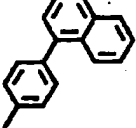
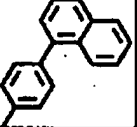
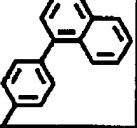
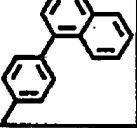
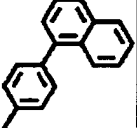








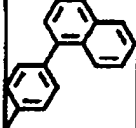
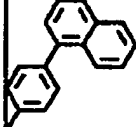
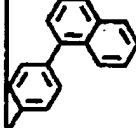
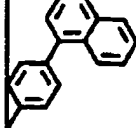
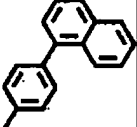
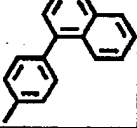
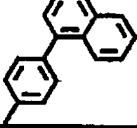
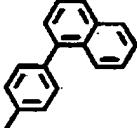




[0044]





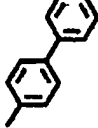
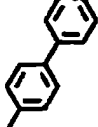
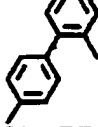
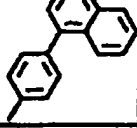
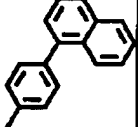
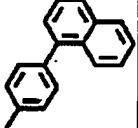
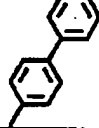
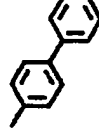
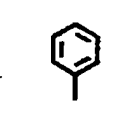







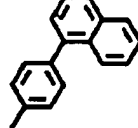
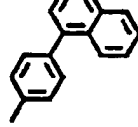
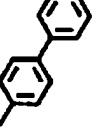
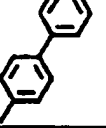
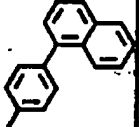
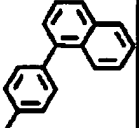
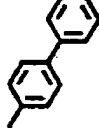
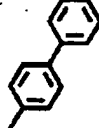








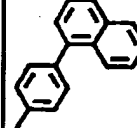
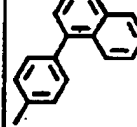
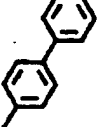
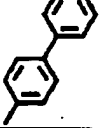
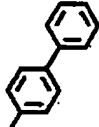
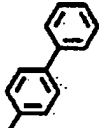
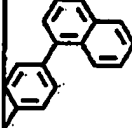
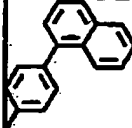




化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>2</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
137									H	H	H	H
138									H	H	H	H
139									CH <sub>3</sub>	CH <sub>3</sub>	H	H
140									OCH <sub>3</sub>	OCH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
141									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
142									OCH <sub>3</sub>	OCH <sub>3</sub>	H	H





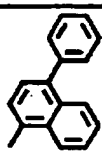
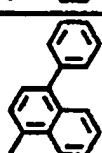
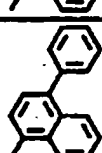
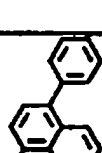
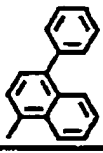
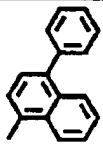
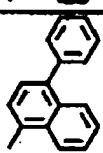
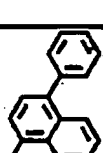








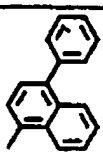
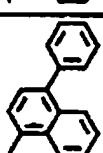
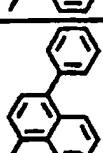
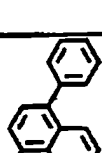
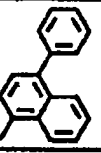
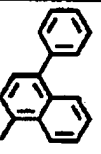
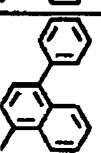
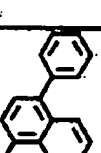








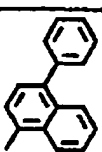
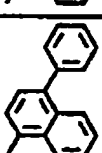
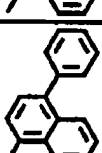
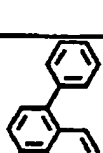
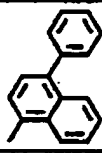
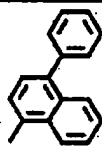
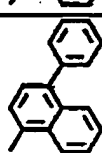
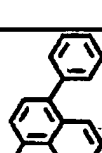
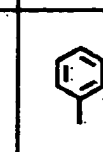



化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
143									H	H	H	H
144									H	H	H	H
145									CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
146									OCH <sub>3</sub>	OCH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
147									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
148									OCH <sub>3</sub>	OCH <sub>3</sub>	H	H

[0046]





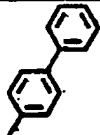
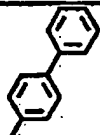
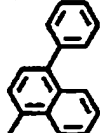
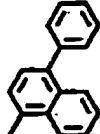
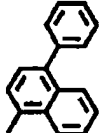
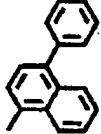
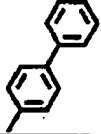
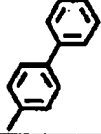








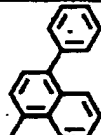
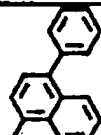
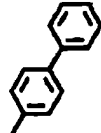
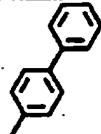
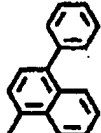
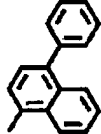
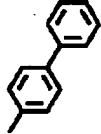
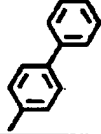








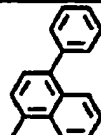
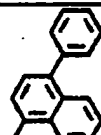
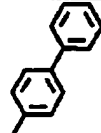
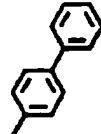
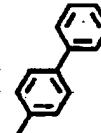
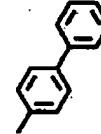
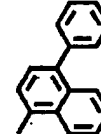
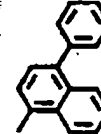




化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
149	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
150	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
151	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								
152	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H								
153	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
154	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
155	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
156	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
157	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								
158	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H								
159	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
160	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								



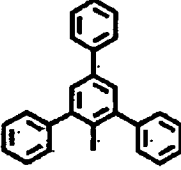
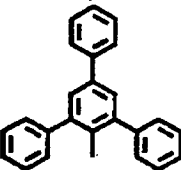










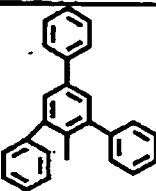
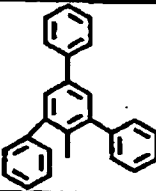


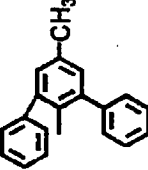
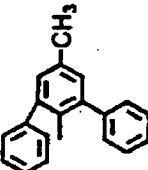










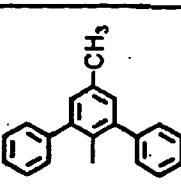
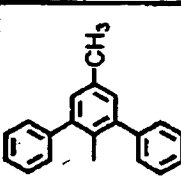


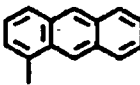
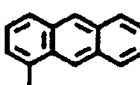




化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
161	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
162	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
163	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								
164	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H								
165	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
166	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
167	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
168	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
169	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								
170	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H								
171	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
172	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								



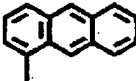
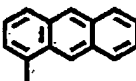


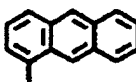
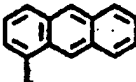


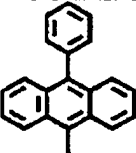
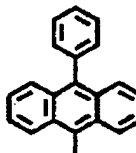






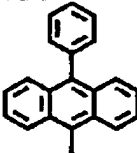
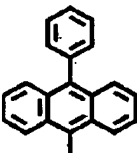






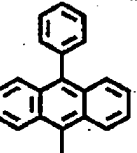
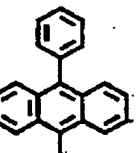


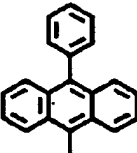
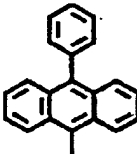


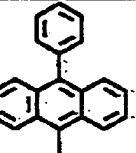
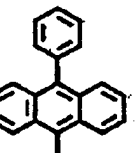




[0050]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
173	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
174	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
175	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								
176	OCH <sub>3</sub> OCH <sub>3</sub>		H	H								
177	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
178	SCH <sub>3</sub> SCH <sub>3</sub>		H	H								



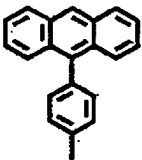
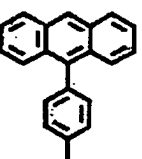


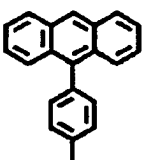
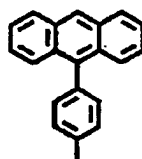


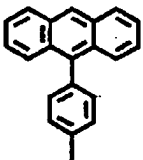
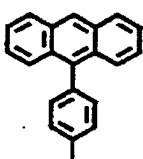






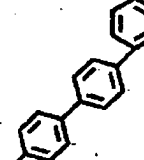
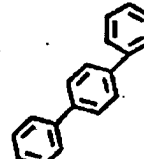






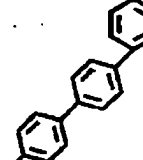
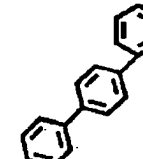


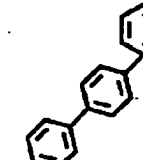
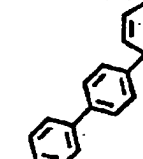


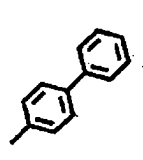
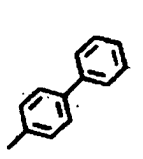


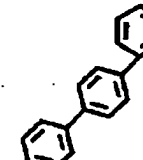
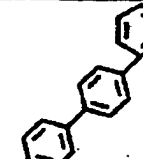


化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
179	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
180	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
181	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
182	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
183	CH <sub>3</sub>	CH <sub>3</sub>	H	H								

[0052]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
184	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
185	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
186	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
187	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
188	CH <sub>3</sub>	CH <sub>3</sub>	H	H								

[0053]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
189	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
190	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
191	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
192	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
193	CH <sub>3</sub>	CH <sub>3</sub>	H	H								

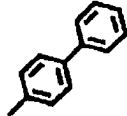
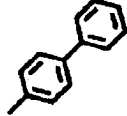
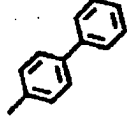
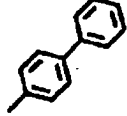





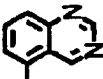
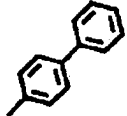
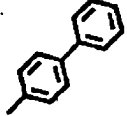






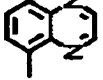
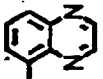
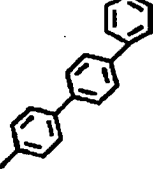
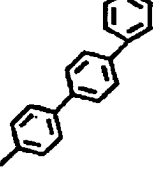






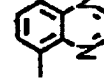
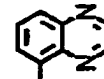
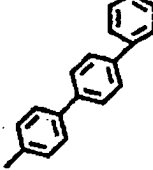
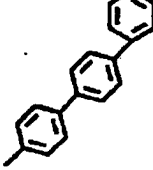
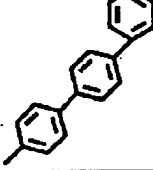
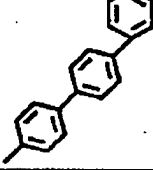




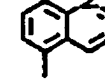
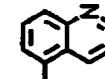
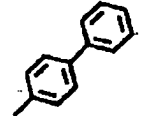
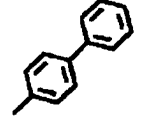
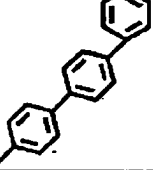
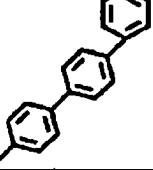




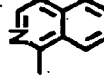

[0054]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>	
194										CH <sub>3</sub>	CH <sub>3</sub>	H	H
195										SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
196										H	OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>
197										CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
198										SCH <sub>3</sub>	SCH <sub>3</sub>	H	H





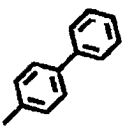
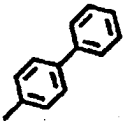


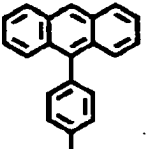
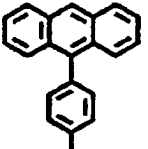




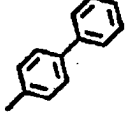
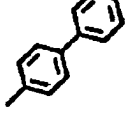




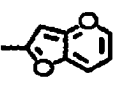
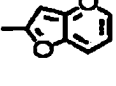


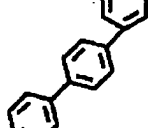
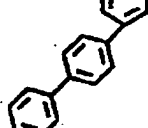








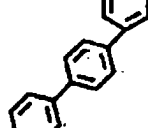
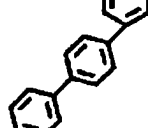


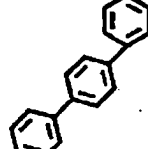
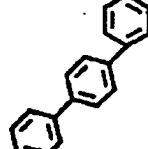




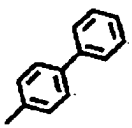
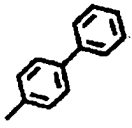


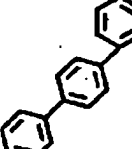
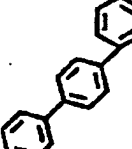
[0055]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
199										CH <sub>3</sub>	H	H
200										SCH <sub>3</sub>	H	H
201										H	OCH <sub>3</sub>	OCH <sub>3</sub>
202										CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
203										SCH <sub>3</sub>	H	H


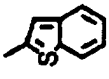


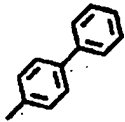
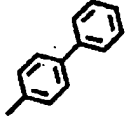


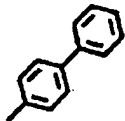
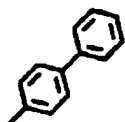




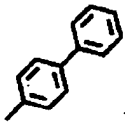
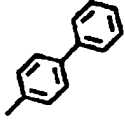








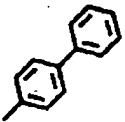
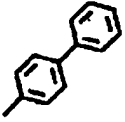








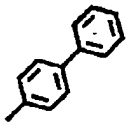
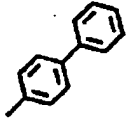


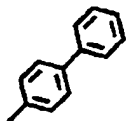
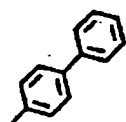




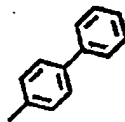
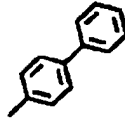


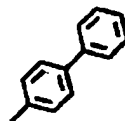
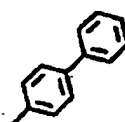
[0056]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
204											H	H
205											H	H
206											H	H
207											H	H
208											H	H

















































[0057]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
209	CH <sub>3</sub>	CH <sub>3</sub>										
210	H	H										
211	SCH <sub>3</sub>	SCH <sub>3</sub>										
212	H	H										
213	CH <sub>3</sub>	CH <sub>3</sub>										

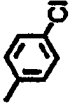
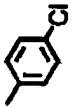














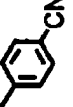
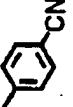






















[0058]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
214	CH <sub>3</sub>	CH <sub>3</sub>										
215	H	H										
216	H	H										
217	H	H										
218	CH <sub>3</sub>	CH <sub>3</sub>										



化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
219	H	H	CN	CN								
220	H	H	Br	Br								
221	H	H	NO <sub>2</sub>	NO <sub>2</sub>								
222	CN	CN	H	H								
223	H	H	Br	Br								
224	Cl	Cl	H	H								

[0060]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
225									H	H	H	H
226									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
227									H	H	CH <sub>3</sub>	CH <sub>3</sub>
228									CH <sub>3</sub>	CH <sub>3</sub>	H	H
229									H	H	H	H

[0061]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
230									H	H	H	H
231									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
232									H	H	CH <sub>3</sub>	CH <sub>3</sub>
234									CH <sub>3</sub>	CH <sub>3</sub>	H	H
235									H	H	H	H

[0062] An organic EL device is a component in which one layer or a multilayer organic compound thin film was formed between an anode plate and cathode. In the case of the mold, the luminous layer is further prepared between an anode plate and cathode. A luminous layer contains luminescent material, and in order to make the electron poured in from the electron hole or cathode which was poured in from the anode plate in addition to it convey to luminescent material, it may contain a hole-injection ingredient, an electron hole transportation ingredient or an electron injection ingredient, and an electronic transportation ingredient. An electron injection

ingredient is an ingredient with the capacity for an electron to be poured in and to get from cathode, and an electronic transportation ingredient is an ingredient with the capacity to convey the poured-in electron to a luminous layer. A hole-injection ingredient is an ingredient with the capacity for an electron hole to be poured in and to get from an anode plate, and an electron hole transportation ingredient is an ingredient with the capacity to convey the poured-in electron hole to a luminous layer.

[0063] As a multilayer mold, there is an organic EL device which carried out the laminating with the multilayer configuration of an anode plate / hole-injection band / luminous layer / cathode, an anode plate / luminous layer / electron injection band / cathode, and an anode plate / hole-injection band / luminous layer / electron injection band / cathode.

[0064] The compound shown by the general formula [1] of this invention is a compound which has strong fluorescence in a solid state, and since it excels in electroluminescence nature, it can be used within a luminous layer as a luminescent material. Moreover, the optimal selection of high luminous efficiency and luminescence wavelength is possible for the compound of a general formula [1] by doping in a luminous layer at a rate that it is the optimal in a luminous layer as a doping ingredient. Furthermore, although carriers, such as an electron hole or an electron, can be conveyed, since electron hole transportability is more excellent, the compound of a general formula [1] can be used as a hole-injection layer. When a hole-injection band is constituted above a bilayer, it can be used for any hole-injection layer.

[0065] The compound of a general formula [1] can be used for the host ingredient of a luminous layer as a doping ingredient (guest ingredient), and an organic EL device with high luminescence brightness can also be obtained. As for the compound of a general formula [1], it is desirable in a luminous layer to contain to a host ingredient in 0.001 % of the weight - 50% of the weight of the range, and 0.01 more % of the weight - 10% of the weight of its range is effective for it.

[0066] As the host ingredient which can be used combining with the compound of a general formula [1], A quinoline metal complex, OKISA diazole, a benzothiazole metal complex, A benzo oxazole metal complex, a benzimidazole metal complex, triazole, An imidazole, oxazole, OKISA diazole, a stilbene, a butadiene, A benzidine mold triphenylamine, a styryl amine mold triphenylamine, diamine mold triphenylamine full -- me -- a non and diamino anthracene mold triphenylamine -- A diamino phenanthrene mold triphenylamine, anthra quinodimethan, There are polymeric materials of conductive polymers, such as diphenquinone, thiadiazole, tetrazole, perylene tetracarboxylic acid, deflection ORENIRIDEN methane, anthra quinodimethan, triphenylene, an anthrone, etc. those derivatives and a polyvinyl carbazole, and polysilane, etc.

[0067] Furthermore, it is also possible to change the luminescent color by using the compound of a general formula [1] as a host ingredient using other doping ingredients. As the doping ingredient used with a general formula [1], An anthracene, naphthalene, a phenanthrene, a pyrene, tetracene, Coronene, a chrysene, a fluorescein, perylene, phtalo perylene, Non [ naphthalo perylene and peri non, / phtalo peri non, / naphthalo peri ] A diphenyl butadiene, a tetra-phenyl butadiene, a coumarin, OKISA diazole, Aldazine, bis-benzo KISAZORIN, bis-styryl, pyrazine, a cyclopentadiene, A quinoline metal complex, an amino quinoline metal complex, an imine, diphenylethylene, Although there are those derivatives, such as a vinyl anthracene, a diamino carbazole, a pyran, thiopyran, poly methine, merocyanine, an imidazole chelation oxy-NOIDO compound, Quinacridone, and rubrene, it is not limited to these.

[0068] In addition to luminescent material and a doping ingredient, if there is need, a hole-injection ingredient and an electron injection ingredient can also be used for a luminous layer.

[0069] An organic EL device can prevent the fall of the brightness by quenching, or a life by making it multilayer structure. Moreover, if there is need, two or more kinds can also be used combining luminescent material, a doping ingredient, and the hole-injection ingredient and electron injection ingredient that performs carrier impregnation. Furthermore, the hole-injection layer, the luminous layer, and the electron injection layer may be formed of the lamination more than a bilayer, respectively, an electron hole or an electron is efficiently poured in from an electrode, and the component structure which may be efficiently conveyed in a layer is chosen.

[0070] What has a bigger work function than 4eV as a conductive ingredient used for the anode plate of an organic EL device is suitable, and organic conductive resin, such as gold oxide groups, such as tin oxide called those alloys and an ITO substrate, and a NESA substrate, indium oxide, etc., such as carbon, aluminum, vanadium, iron, cobalt, nickel, a tungsten, silver, gold, platinum, and palladium, and also the poly thiophene, and polypyrrole, is used.

[0071] What has a work function smaller than 4eV as a conductive ingredient used for cathode is suitable, and those alloys, such as magnesium, calcium, tin, lead, titanium, an yttrium, a lithium, a ruthenium, and manganese, are used. As an alloy, although magnesium/silver, magnesium/indium, a lithium/aluminum, etc. are mentioned as an example of representation, it is not limited to these. The ratio of an alloy is controlled by the temperature of heating, the ambient atmosphere, and the degree of vacuum, and a suitable ratio is chosen. As long as an anode plate and cathode have the need, they may be formed of the lamination more than a bilayer.

[0072] In order to make light emit efficiently in an organic EL device, in the luminescence wavelength field of a component, the transparent enough thing of at least one side is desirable among the anode plates and cathode which are used. Moreover, the transparent thing of a substrate is desirable. The above-mentioned conductive ingredient is used for a transparent electrode, and it sets it up so that predetermined translucency may be secured by approaches, such as vacuum evaporation and sputtering. As for the electrode of a luminescence side, it is desirable to make light transmittance 10% or more.

[0073] A substrate has mechanical and thermal reinforcement, if transparent, it is not limited, but if it illustrates, transparency resin, such as a glass substrate, a polyethylene plate, a polyether ape phon plate, and a polypropylene plate, will be mentioned.

[0074] Formation of each class of the organic EL device concerning this invention is applicable by any approach of the wet forming-membranes methods, such as the dry type forming-membranes methods, such as vacuum deposition and sputtering, and spin coating, dipping. Although especially thickness is not limited, it needs to set each class as suitable thickness. Applied voltage big [ when the film is too thick ] in order to obtain a fixed optical output is needed, effectiveness worsens, if the film is too thin, a pinhole etc. will occur, and even if it impresses an electrical potential difference, sufficient luminescence brightness is not obtained. Usually, although the range of 10 micrometers is suitable from 5nm as thickness used, the range of 0.2 micrometers is still more desirable from 10nm.

[0075] In the case of the wet forming-membranes method, the ingredient which forms each class is dissolved or distributed to suitable solvents, such as chloroform, a tetrahydrofuran, and dioxane, and an organic compound thin film is formed.

[0076] Also in which thin film prepared from dry type or the wet forming-membranes method, resin or an additive suitable for the purposes, such as pinhole prevention, may be used on a membrane formation disposition. As such resin, conductive resin, such as photoconductivity resin, such as insulating resin, such as polystyrene, a polycarbonate, polyarylate, polyester, a polyamide, polyurethane, polysulfone, polymethylmethacrylate, polymethyl acrylate, and a cellulose, Polly N-vinylcarbazole, and polysilane, the poly thiophene, and polypyrrole, can be mentioned. Moreover, an antioxidant, an ultraviolet ray absorbent, a plasticizer, etc. can be mentioned as an additive.

[0077] The compound which has the capacity to pour in an electron hole, as a hole-injection ingredient, has the hole-injection effectiveness which was excellent to a luminous layer or luminescent material, and prevented migration into the electron injection layer or electron injection ingredient of an exciton generated by the luminous layer, and was excellent in thin film organization potency is mentioned. Specifically A phthalocyanine system compound, a naphthalocyanine system compound, A porphyrin system compound, OKISA diazole, triazole, an imidazole, Imidazolone, imidazole thione, pyrazoline, a pyrazolone, a tetrahydro imidazole, Oxazole, OKISA diazole, a hydrazone, an acyl hydrazone, The poly aryl alkane, a stilbene, a butadiene, a benzidine mold triphenylamine, a styryl amine mold triphenylamine, a diamine mold triphenylamine, etc., Although there are polymeric materials, such as those derivatives and a polyvinyl carbazole, polysilane, and a conductive polymer, etc., It is not limited to these.

[0078] The compound which has the capacity to pour in an electron, as an electron injection ingredient, has the electron injection effectiveness which was excellent to a luminous layer or luminescent material, and prevented migration into the hole-injection layer or hole-injection ingredient of an exciton generated by the luminous layer, and was excellent in thin film organization potency is mentioned. as an example -- a quinoline metal complex, OKISA diazole, a benzothiazole metal complex, a benzo oxazole metal complex, a benzimidazole metal complex, and full -- me -- non, although anthra quinodimethan, diphenquinone, thiopyran dioxide, OKISA diazole, thiadiazole, tetrazole, perylene tetracarboxylic acid, deflection ORENIRIDEN methane, anthra quinodimethan, anthrones, etc. those derivatives, etc. are mentioned, it is not limited to these. Moreover, the sensitization by adding the electronic acceptance matter into a hole-injection ingredient, and adding the electron-donative matter into an electron injection ingredient is also possible.

[0079] It is also possible to prepare a protective layer on the surface of a component, or to enclose a silicone oil etc., and to protect the whole component for the improvement of stability to the temperature of the organic EL device obtained by this invention, humidity, an ambient atmosphere, etc.

[0080]

[Example] The example which used the compound of this invention for below is shown. In this example, all mixing ratios are weight ratios. Moreover, the property of an electrode surface product 2mmx2mm organic EL device was measured.

[0081] On the glass plate with an ITO electrode washed example 1, it is compound [ of Table 1 ] (19), 2, and 5-screw (1-naphthyl) as a luminescent material. - 1, 3, 4-OKISA diazole, and polycarbonate resin (Teijin formation : panlight K-1300) were dissolved in the tetrahydrofuran by the weight ratio of 1:2:10, and the luminous layer of 100nm of thickness was obtained with the spin coating method. The electrode of 150nm of thickness was formed with the alloy which moreover mixed silver with magnesium by 10:1 (weight ratio), and the organic EL device was obtained. As for the luminescence property of this component, luminescence of the luminescence brightness 50 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 580 (cd/m<sup>2</sup>), and luminous efficiency 0.62 (lm/W) was obtained.

[0082] On the glass plate with an ITO electrode washed example 2, they are N, N'-(3-methylphenyl)-N, and N'-diphenyl. - Vacuum deposition of 1, the 1'-biphenyl -4, and the 4'-diamine (TPD) was carried out, and the hole-injection layer of 20nm of thickness was obtained. Subsequently, the compound (2) of Table 1 was vapor-deposited, the luminous layer of 40nm of thickness was created, subsequently the tris(8-hydroxyquinolate) aluminium complex (Alq<sub>3</sub>) was vapor-deposited, and the electron injection layer of 30nm of thickness was obtained. The electrode of 100nm of thickness was formed with the alloy which moreover mixed silver with magnesium by 10:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 580 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 13600 (cd/m<sup>2</sup>), and luminous efficiency 1.6 (lm/W) was obtained.

[0083] On the glass plate with an ITO electrode washed example 3, the compound (31) of Table 1 was dissolved in the methylene chloride, and the hole-injection mold luminous layer of 50nm of thickness was obtained with the spin coating method. Subsequently, vacuum deposition of the screw (2-methyl-8-hydroxyquinolate) (1-naphth RATO) gallium complex was carried out, the electron injection layer of 40nm of thickness was created, the electrode of 100nm of thickness was formed on it with the alloy which mixed silver with magnesium by 10:1 (weight ratio), and the organic EL device was obtained. The electron injection layer was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 320 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 9300 (cd/m<sup>2</sup>), and luminous efficiency 1.3 (lm/W) was obtained.

[0084] On the glass plate with an ITO electrode washed example 4, vacuum deposition of the compound (41) of Table 1 was carried out, and the hole-injection mold luminous layer of 50nm of thickness was obtained. Subsequently, vacuum deposition of the screw (2-methyl-8-hydroxyquinolate) (p-cyano phenolate) gallium complex was carried out, the electron injection layer of 30nm of thickness was created, the electrode of 100nm of thickness was formed on it with the alloy which mixed silver with magnesium by 10:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 390 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 13900 (cd / m<sup>2</sup>), and luminous efficiency 1.4 (lm/W) was obtained.

[0085] On the glass plate with an ITO electrode washed example 5-19, vacuum deposition of the 4 and 4' screw [ - ] [N-(1-naphthyl)-N-phenylamino] biphenyl (alpha-NPD) was carried out, and the hole-injection layer of 30nm of thickness was formed. Subsequently, vacuum deposition of the ingredient shown in Table 2 as a luminescent material was carried out, and the luminous layer of 30nm of thickness was obtained. Subsequently, vacuum deposition of the screw (2-methyl-8-hydroxyquinolate) (phenolate) gallium complex was carried out, the electron injection layer of 30nm of thickness was created, the electrode of 100nm of thickness was formed on it with the alloy which mixed silver with magnesium by 10:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. The luminescence property of this component is shown in Table 2. Luminescence brightness here is brightness at the time of direct-current-voltage 5V seal of approval. All the organic EL

devices of this example had the high brightness property more than highest brightness 10000 (cd/m<sup>2</sup>).

[0086] Table 2 [Table 2]

【表2】

実施例	表1の発光材料	発光輝度 (cd/m <sup>2</sup> )	最大発光輝度 (cd/m <sup>2</sup> )	最大発光効率 (lm/W)
5	(4)	290	17500	1.4
6	(40)	610	19700	1.8
7	(45)	690	18700	1.5
8	(92)	140	12900	1.7
9	(96)	350	17200	1.3
10	(101)	550	17300	2.0
11	(103)	320	12600	1.5
12	(104)	520	19900	1.6
13	(111)	810	11300	1.4
14	(133)	420	11200	2.0
15	(151)	770	11500	1.3
16	(168)	420	18200	1.7
17	(179)	250	10700	1.0
18	(194)	290	18300	1.8
19	(209)	730	16900	1.3
27	(81)	530	18200	2.1
28	(90)	510	26300	2.3
29	(102)	610	27900	2.6
30	(137)	670	19900	1.9
31	(197)	420	20600	2.5

発光輝度は直流5V印加時の値

[0087] On the glass plate with an ITO electrode washed example 20, vacuum deposition of 4, 4', and the 4"-tris [N-(3-methylphenyl)-N-phenylamino] triphenylamine was carried out, and the hole-injection layer of 40nm of thickness was obtained. Subsequently, vacuum deposition of alpha-NPD was carried out, and the second hole-injection layer of 10nm of thickness was obtained. Furthermore, vacuum deposition of the compound (17) of Table 1 was carried out, the luminous layer of 30nm of thickness was created, vacuum deposition of the screw (2-methyl-8-hydroxyquinolate) (phenolate) gallium complex was carried out further, the electron injection layer of 30nm of thickness was created, the electrode of 150nm of thickness was formed on it with the alloy which mixed aluminum and a lithium by 25:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 840 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 13200 (cd/m<sup>2</sup>), and luminous efficiency 1.9 (lm/W) was obtained.

[0088] On the glass plate with an ITO electrode washed example 21, vacuum deposition of the 4 and 4' screw [ - ] [N-(1-naphthyl)-N-phenylamino] biphenyl (alpha-NPD) was carried out, and the hole-injection layer of 20nm of thickness was obtained. Subsequently, vacuum deposition of the compound (26) of Table 1 was carried out, the luminous layer of 40nm of thickness was created, subsequently the tris(8-hydroxyquinolate) aluminium complex (Alq3) was vapor-deposited, and the electron injection layer of 30nm of thickness was obtained. 0.5nm of electrodes was first formed [ lithium fluoride (LiF) ] further for aluminum (aluminum) with 200nm vacuum deposition on it, and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 300 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 17400 (cd/m<sup>2</sup>), and luminous efficiency 1.9 (lm/W) was obtained.

[0089] The organic EL device was produced by the same approach as an example 3 except preparing the hole-injection layer of 5nm of thickness of a copper phthalocyanine between an example 22ITO electrode and the compound (201) of Table 1. As for this component, luminescence of the luminescence brightness 580 (cd/m<sup>2</sup>), the maximum luminescence brightness 16200 (cd/m<sup>2</sup>), and luminous efficiency 1.7 (lm/W) was obtained by direct-current-voltage 5V.

[0090] an example 234, 4', and 4" -- the organic EL device was produced by the same approach as an example 20 except preparing the hole-injection layer of 20nm of thickness of a non-metal phthalocyanine instead of - tris

[N-(3-methylphenyl)-N-phenylamino] triphenylamine. As for this component, luminescence of the luminescence brightness 560 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 21100 (cd/m<sup>2</sup>), and luminous efficiency 2.1 (lm/W) was obtained.

[0091] The organic EL device was produced by the same approach as an example 5 except preparing the compound (13) of Table 1, and the thin film of 30nm of thickness which vapor-deposited (15) by the weight ratio of 2:3 as example 24 luminous layer. This component is the luminescence brightness 900 (cd/m<sup>2</sup>) maximum luminescence by direct-current-voltage 5V. Luminescence of brightness 21700 (cd/m<sup>2</sup>) and luminous efficiency 2.5 (lm/W) was obtained.

[0092] The organic EL device was produced by the same approach as an example 5 except preparing the thin film of 30nm of thickness which vapor-deposited the compound (24) and tris(8-hydroxyquinolate)aluminium complex (Alq<sub>3</sub>) of Table 1 by the weight ratio of 1:10 as example 25 luminous layer. As for this component, luminescence of the luminescence brightness 360 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 18900 (cd / m<sup>2</sup>), and luminous efficiency 2.2 (lm/W) was obtained.

[0093] The organic EL device was produced by the same approach as an example 5 except preparing the compound (68) of Table 1, and the thin film of 30nm of thickness which vapor-deposited the 4 and 4'-screw (beta and beta-diphenyl vinyl) biphenyl by the weight ratio of 3:100 as example 26 luminous layer. As for this component, luminescence of the luminescence brightness 440 (cd/m<sup>2</sup>) maximum luminescence brightness 23500 (cd/m<sup>2</sup>) of direct-current-voltage 5V and luminous efficiency 2.5 (lm/W) was obtained.

[0094] The organic EL device was produced by the same approach as an example 5 except preparing the thin film of the compound shown in Table 2, and 30nm of thickness which vapor-deposited 4 and 4' screw [ - ] [N-(9-phenan tolyl)-N-phenylamino] biphenyl by the weight ratio of 1:20 as 27 to example 31 luminous layer. The luminescence property of this component is shown in Table 2. Luminescence brightness here is brightness at the time of direct-current-voltage 5V seal of approval. All the organic EL devices of this example had the high brightness property more than highest brightness 15000 (cd/m<sup>2</sup>).

[0095] The organic EL device was produced by the same approach as an example 5 except preparing the compound (227) of Table 1, and the thin film of 30nm of thickness which vapor-deposited the 4-(dicyanomethylene)-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM) by the weight ratio of 100:3 as example 32 luminous layer. This component is the luminescence brightness 880 (cd/m<sup>2</sup>) maximum luminescence by direct-current-voltage 5V. Luminescence of brightness 15000 (cd/m<sup>2</sup>) and luminous efficiency 1.8 (lm/W) was obtained.

[0096] On the glass plate with an ITO electrode washed example 33, vacuum deposition of the copper phthalocyanine was carried out, and the hole-injection layer of 50nm of thickness was obtained. Subsequently, vacuum deposition of alpha-NPD was carried out, and the second hole-injection layer of 30nm of thickness was obtained. Furthermore, vacuum deposition of the compound (73) of Table 1 was carried out, the luminous layer of 30nm of thickness was created, vacuum deposition of the tris(8-hydroxyquinolate)aluminium complex was carried out further, the electron injection layer of 30nm of thickness was created, the electrode of 150nm of thickness was formed on it with the alloy which mixed aluminum and a lithium by 25:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 740 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 18700 (c d/m<sup>2</sup>), and luminous efficiency 2.4 (lm/W) was obtained. Moreover, when carrying out continuation luminescence of this component by 3 (mA/cm<sup>2</sup>), luminescence stabilized for 3000 hours or more has been observed.

[0097] On the glass plate with an ITO electrode washed example 34, the compound (19) of Table 1 was dissolved in the methylene chloride, and the hole-injection layer of 50nm of thickness was obtained with the spin coating method. Subsequently, the screw (2-methyl-8-hydroxyquinolate) (phenolate) gallium complex was vapor-deposited, and the electron injection mold luminous layer of 70nm of thickness was obtained. The electrode of 100nm of thickness was formed with the alloy which mixed silver with magnesium by 10:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 940 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 15900 (cd/m<sup>2</sup>), and luminous efficiency 1.7 (lm/W) was obtained.

[0098] On the glass plate with an ITO electrode washed example 35, vacuum deposition of the copper



phthalocyanine was carried out, and the hole-injection layer of 50nm of thickness was obtained. Subsequently, vacuum deposition of the compound (49) of Table 1 was carried out, and the second hole-injection layer of 30nm of thickness was obtained. Furthermore, vacuum deposition of N, N, N', N'-tetrakis [p-(alpha and alpha-dimethylbenzyl) phenyl]-9, and the 10-anthracene diamine is carried out, The luminous layer of 30nm of thickness was created, vacuum deposition of the tris(8-hydroxyquinolate)aluminium complex was carried out further, the electron injection layer of 30nm of thickness was created, the electrode of 150nm of thickness was formed on it with the alloy which mixed aluminum and a lithium by 25:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 670 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 24500 (c d/m<sup>2</sup>), and luminous efficiency 2.6 (lm/W) was obtained.

[0099] On the glass plate with an ITO electrode washed example 36, vacuum deposition of the compound (57) of Table 1 was carried out, and the hole-injection layer of 30nm of thickness was obtained. Subsequently, vapor codeposition of the rubrene was carried out to alpha-NPD by the weight ratio 100:3, the luminous layer of 40nm of thickness was created, subsequently the tris(8-hydroxyquinolate)aluminium complex (Alq<sub>3</sub>) was vapor-deposited, and the electron injection layer of 30nm of thickness was obtained. 0.5nm of electrodes was first formed [ lithium fluoride (LiF) ] further for aluminum (aluminum) with 200nm vacuum deposition on it, and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 740 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 16400 (cd/m<sup>2</sup>), and luminous efficiency 1.7 (lm/W) was obtained.

[0100] On the glass plate with an ITO electrode washed example 37, vacuum deposition of the compound (65) of Table 1 was carried out, and the hole-injection layer of 40nm of thickness was obtained. Subsequently, vacuum deposition of alpha-NPD was carried out, and the second hole-injection layer of 10nm of thickness was obtained. Furthermore, carry out vapor codeposition of tris(8-hydroxyquinolate)aluminium and the 3-(2'-benzothiazolyl)-7-diethylamino coumarin (coumarin 6) by the weight ratio 100:2. The luminous layer of 30nm of thickness was created, vacuum deposition of the screw (2-methyl-8-hydroxyquinolate) (phenolate) gallium complex was carried out further, the electron injection layer of 30nm of thickness was created, the electrode of 150nm of thickness was formed on it with the alloy which mixed aluminum and a lithium by 25:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 860 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 28100 (c d/m<sup>2</sup>), and luminous efficiency 2.3 (lm/W) was obtained.

[0101] On the glass plate with an ITO electrode washed example 38, vacuum deposition of the compound (199) of Table 1 was carried out, and the hole-injection layer of 40nm of thickness was obtained. Subsequently, vacuum deposition of the compound (49) of Table 1 was carried out, and the second hole-injection layer of 10nm of thickness was obtained. Furthermore, vacuum deposition of the 4 and 4'-screw (beta and beta-diphenyl vinyl) biphenyl was carried out, the luminous layer of 30nm of thickness was created, vacuum deposition of the screw (2-methyl-8-hydroxyquinolate) (phenolate) gallium complex was carried out further, the electron injection layer of 30nm of thickness was created, the electrode of 100nm of thickness was formed on it with the alloy which mixed silver with magnesium by 10:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 380 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 24300 (c d/m<sup>2</sup>), and luminous efficiency 2.4 (lm/W) was obtained.

[0102] On the glass plate with an ITO electrode washed example 39, vacuum deposition of the 4 and 4' screw [ - ] [N-(1-naphthyl)-N-phenylamino] biphenyl (alpha-NPD) was carried out, and the hole-injection layer of 30nm of thickness was formed. Subsequently, vapor codeposition of tris(8-hydroxyquinolate)aluminium and the 3-(2'-benzothiazolyl)-7-diethylamino coumarin (coumarin 6) was carried out by the weight ratio 100:2, and the luminous layer of 30nm of thickness was obtained. Subsequently, vacuum deposition of the compound (204) of Table 1 was carried out, the electron injection layer of 30nm of thickness was created, the electrode of 150nm of thickness was formed on it with the alloy which mixed aluminum and a lithium by 25:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a

substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 290 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 19700 (cd / m<sup>2</sup>), and luminous efficiency 2.6 (lm/W) was obtained.

[0103] On the glass plate with an ITO electrode washed example 40, vacuum deposition of 2, 3, 6, 7, 10, and the 11-hexamethoxy triphenylene was carried out, and the hole-injection layer of 30nm of thickness was formed. subsequently, N, N, N', and N' – vacuum deposition of - tetra-p-biphenyl -1 and the 4-naphthalene diamine was carried out, and the luminous layer of 30nm of thickness was obtained. Subsequently, vacuum deposition of the compound (217) of Table 1 was carried out, the second electron injection layer of 10nm of thickness was created, vacuum deposition of the screw (2-methyl-8-hydroxyquinolate) (phenolate) gallium complex was carried out further, and the first electron injection layer of 30nm of thickness was created. The electrode of 100nm of thickness was formed with the alloy which moreover mixed silver with magnesium by 10:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 760 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 25400 (cd / m<sup>2</sup>), and luminous efficiency 2.9 (lm/W) was obtained.

[0104] On the glass plate with an ITO electrode washed example 41, vacuum deposition of the 4 and 4' screw [ - ] [N-(9-phenan tolyl)-N-phenylamino] biphenyl was carried out, and the hole-injection layer of 30nm of thickness was formed. Subsequently, vapor codeposition of N and N'-dimethyl Quinacridone was carried out to the compound (235) of Table 1 by the weight ratio 150:1, and the luminous layer of 40nm of thickness was obtained. Subsequently, vacuum deposition of the compound (227) of Table 1 was carried out, and the electron injection layer of 30nm of thickness was created. The electrode of 100nm of thickness was formed with the alloy which moreover mixed silver with magnesium by 10:1 (weight ratio), and the organic EL device was obtained. Each class was vapor-deposited under the conditions of a substrate temperature room temperature in the vacuum of ten to 4 Pa. As for this component, luminescence of the luminescence brightness 710 (cd/m<sup>2</sup>) of direct-current-voltage 5V, the maximum luminescence brightness 35000 (cd / m<sup>2</sup>), and luminous efficiency 4.6 (lm/W) was obtained.

[0105] The organic EL device was produced by the same approach as an example 5 except preparing the thin film of 30nm of thickness which carried out vacuum deposition of 5, 6, 11, and the 12-tetra-phenyl naphthacene (rubrene) as example of comparison 1 luminous layer. This component was luminescence of the luminescence brightness 50 (cd/m<sup>2</sup>) maximum luminescence brightness 1880 (cd/m<sup>2</sup>) of direct-current-voltage 5V, and luminous efficiency 0.3 (lm/W).

[0106] As example of comparison 2 luminous layer, they are 5, 6, 11, and 12-tetra-biphenyl - The organic EL device was produced by the same approach as an example 20 except preparing the thin film of 30nm of thickness which carried out vacuum deposition of the 2, 3, 8, and 9-tetra-phenyl naphthacene. This component was luminescence of the luminescence brightness 270 (cd/m<sup>2</sup>) maximum luminescence brightness 2400 (cd/m<sup>2</sup>) of direct-current-voltage 5V, and luminous efficiency 0.25 (lm/W).

[0107] The organic EL device was produced by the same approach as an example 27 except preparing the thin film of 30nm of thickness which vapor-deposited 5, 6, 11, a 12-tetra-biphenyl naphthacene and 4, and 4' screw [ - ] [N-(9-phenan tolyl)-N-phenylamino] biphenyl by the weight ratio of 1:20 as example of comparison 3 luminous layer. This component was luminescence of the luminescence brightness 340 (cd/m<sup>2</sup>) maximum luminescence brightness 7200 (cd/m<sup>2</sup>) of direct-current-voltage 5V, and luminous efficiency 0.95 (lm/W).

[0108] The organic EL device was produced by the same approach as an example 32 except preparing the thin film of 30nm of thickness which vapor-deposited 2, 3, 5, 6, 8, 9, 11, 12-OKUTA phenyl naphthacene, and DCM by the weight ratio of 100:3 as example of comparison 4 luminous layer. This component was luminescence of the luminescence brightness 170 (cd/m<sup>2</sup>) maximum luminescence brightness 5400 (cd/m<sup>2</sup>) of direct-current-voltage 5V, and luminous efficiency 0.85 (lm/W).

[0109] In the component configuration more than a bilayer mold, luminescence more than maximum luminescence brightness 10000 (cd/m<sup>2</sup>) was obtained, and the organic EL device shown by this example was able to acquire high luminous efficiency altogether. About the organic EL device shown by this example, when carrying out continuation luminescence by 3 (mA/cm<sup>2</sup>), luminescence stable for 1000 hours or more was able to be observed.

[0110] The organic EL device of this invention attains improvement in luminous efficiency and luminescence brightness, and reinforcement, and does not limit the component production approaches used collectively, such

as luminescent material, a doping ingredient, a hole-injection ingredient, an electron injection ingredient, a sensitizer, resin, and an electrode material.

[0111]

[Effect of the Invention] The organic EL device which used for the charge of organic EL device material of this invention, and was created is high brightness in high luminous efficiency compared with the former, and has a long luminescence life. When it is used especially for luminescent material, the component of a very high property can be created and it can be suitably used as flat-panel displays and flat-surface emitters, such as a flat TV. Therefore, the application to the light source of the light source of a copying machine, a printer, etc., a liquid crystal display, instruments, etc., the plotting board, a beacon light, etc. is possible. A long lasting organic EL device can be obtained.

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[Translation done.]

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(54) 【発明の名称】 有機エレクトロルミネッセンス素子材料およびそれを使用した有機エレクトロルミネッセンス素子

(57) 【要約】

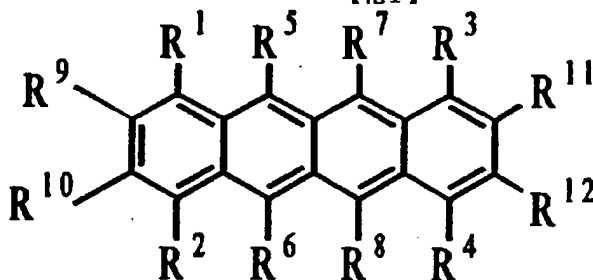
発光効率が高く、繰り返し使用時での安定性の優れた有機EL素子の提供すること。

【課題】

【解決手段】 下記一般式〔1〕で示される有機エレクトロルミネッセンス素子材料。

一般式〔1〕

【化1】



[但し、R<sup>1</sup>~R<sup>12</sup>の内の少なくとも7個はアリール基であり、かつ、R<sup>1</sup>~R<sup>4</sup>が全て同時に水素原子となるこ

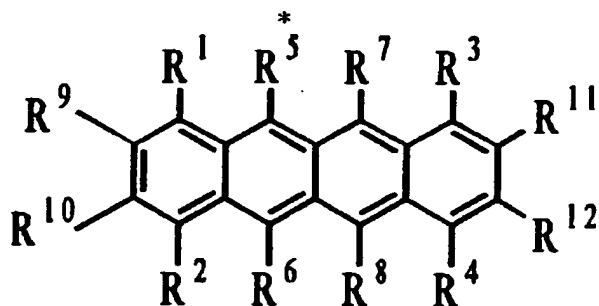
とはない。]

## 【特許請求の範囲】

【請求項1】下記一般式〔1〕で示される有機エレクトロルミネッセンス素子材料。

\*一般式〔1〕

〔化1〕



【但し、式中  $R^1 \sim R^{12}$  はそれぞれ独立に水素原子、ハロゲン原子、または、アルキル基、アリール基、アルコキシ基、アリールオキシ基、アルキルチオ基、アリールチオ基、アミノ基および複素環残基からなる群より選ばれる置換もしくは未置換の有機残基を表し、 $R^1 \sim R^{12}$  は近接した置換基同志で結合して新たな環を形成してもよい。また、 $R^1 \sim R^{12}$  の内の少なくとも7個は、置換もしくは未置換のアリール基であり、かつ、 $R^1 \sim R^4$  が

【請求項2】 一對の電極間に発光層を含む少なくとも一層の有機化合物薄膜を形成した有機エレクトロルミネッセンス素子において、少なくとも一層が請求項1記載の有機エレクトロルミネッセンス素子材料を含有する層である有機エレクトロルミネッセンス素子。

【請求項3】 一對の電極間に発光層を含む複数層の有機化合物薄膜を形成した有機エレクトロルミネッセンス素子において、発光層が請求項1記載の有機エレクトロルミネッセンス素子材料を含有する層である有機エレクトロルミネッセンス素子。

【請求項4】 一對の電極間に発光層を含む複数層の有機化合物薄膜を形成した有機エレクトロルミネッセンス素子において、発光層と陽極との間の正孔注入帯域中の少なくとも一層が請求項1記載の有機エレクトロルミネッセンス素子材料を含有する層である有機エレクトロルミネッセンス素子。

## 【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は平面光源や表示に使用される有機エレクトロルミネッセンス（EL）素子に関するものである。

【0002】

【従来の技術】有機物質を使用したEL素子は、固体発光型の安価な大面積フルカラー表示素子としての用途が有望視され、多くの開発が行われている。一般にEL素子は、発光層および該層をはさんだ一對の対向電極から構成されている。

【0003】発光は、両電極間に電界が印加されると、※

※陰極側から電子が注入され、陽極側から正孔が注入される。さらに、この電子が発光層において正孔と再結合し、エネルギー準位が伝導帯から価電子帯に戻る際にエネルギーを光として放出する現象である。

【0004】従来の有機EL素子は、無機EL素子に比べて駆動電圧が高く、発光輝度や発光効率も低かった。また、特性劣化も著しく実用化には至っていなかった。

【0005】近年、10V以下の低電圧で発光する高い蛍光量子効率を持った有機化合物を含有した薄膜を積層した有機EL素子が報告され、関心を集めている（アプライド・フィジクス・レターズ、51巻、913ページ、1987年参照）。

【0006】この方法では、金属キレート錯体を蛍光体層、アミン系化合物を正孔注入層に使用して、高輝度の緑色発光を得ており、6～7Vの直流電圧で輝度は100cd/m<sup>2</sup>、最大発光効率は1.5lm/Wを達成して、実用領域に近い性能を持っている。しかしながら、現在までの有機EL素子は、構成の改善により発光強度は改良されているが、未だ十分な発光輝度は有していない。また、繰り返し使用時の安定性に劣るという大きな問題を持っている。

【0007】

【発明が解決しようとする課題】本発明の目的は、発光効率が高く、繰り返し使用時での安定性の優れた有機EL素子の提供にある。本発明者らが鋭意検討した結果、一般式〔1〕で示される化合物の有機EL素子材料を少なくとも一層に使用した有機EL素子の発光効率が高く、繰り返し使用時での安定性も優れていることを見だし本発明に至った。

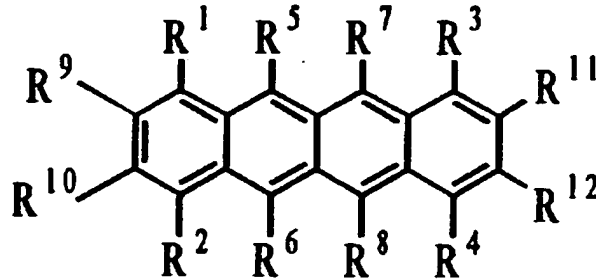
【0008】

【課題を解決するための手段】即ち、本発明は、下記一般式〔1〕で示される有機エレクトロルミネッセンス素子材料に関する。

【0009】一般式〔1〕

【0010】

〔化2〕



【0011】〔但し、式中  $R^1 \sim R^{12}$  はそれぞれ独立に水素原子、ハロゲン原子、または、アルキル基、アリール基、アルコキシ基、アリールオキシ基、アルキルチオ基、アリールチオ基、アミノ基および複素環残基からなる群より選ばれる置換もしくは未置換の有機残基を表し、 $R^1 \sim R^{12}$  は近接した置換基同志で結合して新たな環を形成してもよい。また、 $R^1 \sim R^{12}$  の内の少なくとも7個は、置換もしくは未置換のアリール基であり、かつ、 $R^1 \sim R^4$  が全て同時に水素原子となることはない。〕

また、本発明は一对の電極間に発光層を含む少なくとも一層の有機化合物薄膜を形成した有機エレクトロルミネッセンス素子において、少なくとも一層が上記有機エレクトロルミネッセンス素子材料を含有する層である有機エレクトロルミネッセンス素子に関する。

【0012】また、本発明は一对の電極間に発光層を含む複数層の有機化合物薄膜を形成した有機エレクトロルミネッセンス素子において、発光層が上記有機エレクトロルミネッセンス素子材料を含有する層である有機エレクトロルミネッセンス素子に関する。

【0013】また、本発明は一对の電極間に発光層を含む複数層の有機化合物薄膜を形成した有機エレクトロルミネッセンス素子において、発光層と陽極との間の正孔注入帯域中の少なくとも一層が上記有機エレクトロルミネッセンス素子材料を含有する層である有機エレクトロルミネッセンス素子に関する。

【0014】

【発明の実施の形態】本発明の一般式〔1〕で表される化合物中の  $R^1 \sim R^{12}$  は、それぞれ独立に水素原子、ハロゲン原子、または、下記に示す置換もしくは未置換の有機残基を表す。

【0015】本発明の有機残基とは、アルキル基、アリール基、アルコキシ基、アリールオキシ基、アルキルチオ基、アリールチオ基、アミノ基および複素環残基が挙げられる。

【0016】本発明におけるアリール基としては、フェニル基、ビフェニル基、ターフェニル基、クォーターフェニル基、*o*-、*m*-、および *p*-トリル基、キシリル基、*o*-、*m*-、および *p*-クメニル基、メシチル基、ペンタレニル基、インデニル基、ナフチル基、ビナフタレニル基、ターナフタレニル基、クォーターナフタレニル基、

10 \*ル基、アズレニル基、ヘプタレニル基、ビフェニレニル基、インダセニル基、フルオランテニル基、アセナフチレニル基、アセアントリレニル基、フェナレニル基、フルオレニル基、アントリル基、ビアントラセニル基、ターアントラセニル基、クォーターアントラセニル基、アントラキノリル基、フェナントリル基、トリフェニレニル基、ビレニル基、クリセニル基、ナフタセニル基、アレイアデニル基、ビセニル基、ペリレニル基、ペンタフェニル基、ペンタセニル基、テトラフェニレニル基、ヘキサフェニル基、ヘキサセニル基、ルビセニル基、コロネニル基、トリナフチレニル基、ヘプタフェニル基、ヘプタセニル基、ピラントレニル基、オバレニル基等がある。

【0017】また、本発明における複素環残基としては、チエニル基、ベンゾ〔b〕チエニル基、ナフト〔2, 3-b〕チエニル基、チアントレニル基、フリル基、ピラニル基、イソベンゾフラニル基、クロメニル基、キサンテニル基、フェノキサチニル基、2H-ピロリル基、ピロリル基、イミダゾリル基、ピラゾリル基、ヒリジル基、ピラジニル基、ヒリミジニル基、ヒリダジニル基、インドリジニル基、イソインドリル基、3H-インドリル基、インドリル基、1H-インダゾリル基、アリニル基、4H-キノリジニル基、イソキノリル基、キノリル基、フタラジニル基、ナフチリジニル基、キノキサニル基、キナゾリニル基、シンノリニル基、アテリジニル基、4aH-カルバゾリル基、カルバゾリル基、 $\beta$ -カルボリニル基、フェナントリジニル基、アクリジニル基、ペリミジニル基、フェナントロリニル基、フェナジニル基、フェナルサジニル基、イソチアゾリル基、フェノチアジニル基、イソキサゾリル基、フラザニル基、フェノキサジニル基、イソクロマニル基、クロマニル基、ピロリジニル基、ピロリニル基、イミダゾリジニル基、イミダゾリニル基、ピラゾリジニル基、ピラゾリニル基、ビペリジニル基、ビペラジニル基、インドリニル基、イソインドリニル基、キヌクリジニル基、モルホリニル基等がある。

【0018】また、本発明におけるアルキル基としては、メチル基、エチル基、*n*-および *iso*-プロピル基、*n*-、*iso*-、*sec*-、および *tert*-ブチル基、*n*-、*iso*-、*neo*-、および *tert*-ペンチル基、*n*-、*iso*-、および *neo*-ヘキシル基、ベンジル基等がある。

【0019】本発明におけるアミノ基としては、1級アミノ基、モノアルキルアミノ基、モノアリールアミノ基、モノ複素環残基アミノ基、ジアルキルアミノ基、ジアリールアミノ基、ジ複素環残基アミノ基、アルキルアリールアミノ基、アルキル複素環残基アミノ基、アリール複素環残基アミノ基が挙げられる。

【0020】一般式〔1〕で表される化合物中、 $R^1 \sim R^{12}$ の有機残基は、他の置換基に置換されていても構わない。置換基の種類としては、ハロゲン原子、シアノ基、ニトロ基、置換もしくは未置換のアルキル基、置換もしくは未置換のアルコキシ基、置換もしくは未置換のアルキルチオ基、置換もしくは未置換のアミノ基、置換もしくは未置換のアルキルアミノ基がある。

【0021】本発明において、一般式〔1〕で表される化合物は、例えば次のような方法で合成することが出来る。なお、一般式〔1〕で表される化合物の合成法は、これらに限定されるものではない。

【0022】イソベンゾフラン誘導体をジプロモエチレンと反応させ、ジプロモナフタレン誘導体を合成する。このジプロモナフタレン誘導体をイソベンゾフラン誘導体と反応させ、一般式〔1〕で表される化合物を合成する。

【0023】

【実施例】1, 4, 5, 6, 7, 10, 11, 12, 13-オクタフェニルナフタセンの合成法

2, 3, 6, 7-テトラフェニルイソベンゾフラン3 g(7.1 mmol)と1, 2-ジプロモエチレン2.6 g(14.2 mmol)とp-トルエンスルホン酸50 mgとを、キシレン15 ml中で12時\*

\*間加熱環流した。その後、メタノール15 mlを加え、析出した固体を濾取し、ベンゼン-エタノールで再結晶することで2, 3-ジプロモ-1, 4, 5, 8-テトラフェニルナフタレン2.8 gを得た(収率67%)。次に、2, 3-ジプロモ-1, 4, 5, 8-テトラフェニルナフタレン6.8 g(11.5 mmol)と2, 3, 6, 7-テトラフェニルイソベンゾフラン4.7 g(11.1 mmol)とをテトラヒドロフラン100 mlに溶解し、-78℃でn-ブチルリチウム0.73 g(11.4 mmol)を滴下し、室温まで加温し、12時間攪拌する。反応終了後、水100 mlを加え、さらにヘキサン100 mlを加えて0℃まで冷却して、析出した固体を濾取する。この析出固体をベンゼン-エタノールで再結晶することで5, 12-エポキシ-1, 4, 5, 6, 7, 10, 11, 12-オクタフェニルナフタセン8.2 gを得た(収率89%)。次に、5, 12-エポキシ-1, 4, 5, 6, 7, 10, 11, 12-オクタフェニルナフタセン8.5 g(10.2 mmol)をクロロホルム50 mlに溶解し、-78℃で臭化アルミニウム0.4 g(3.75 mmol)を含むジプロモメタン溶液3.75 mlを滴下し、室温まで加温する。反応終了後、5%二亜硫酸ナトリウム水溶液25 mlを加え、有機層を分離し、硫酸マグネシウムで乾燥を行う。有機層をエバポレーションし、残留物をベンゼン-エタノールで再結晶することで、1, 4, 5, 6, 7, 10, 11, 12, 13-オクタフェニルナフタセン6.6 gを得た(収率80%)。

【0024】本発明の化合物の代表例を表1に具体的に例示するが、本発明は以下の代表例に限定されるものではない。

【0025】表1

【表1】

化合物	$R^1$	$R^2$	$R^3$	$R^4$	$R^5$	$R^6$	$R^7$	$R^8$	$R^9$	$R^{10}$	$R^{11}$	$R^{12}$
1	H								H	H	H	H
2									H	H	H	H
3					H				H	H	H	H
4												
5											H	H
6			H	H								
7		H	H	H							H	H
8		H	H	H						H	H	

【0026】

7

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
9			H	H					H	H		
10			H	H							H	H
11									CH <sub>3</sub>	CH <sub>3</sub>	H	H
12									CH <sub>3</sub>	H	CH <sub>3</sub>	H
13									CH <sub>3</sub>	H	H	CH <sub>3</sub>
14									OCH <sub>3</sub>	OCH <sub>3</sub>	H	H
15									OCH <sub>3</sub>	H	OCH <sub>3</sub>	H
18									OCH <sub>3</sub>	H	H	OCH <sub>3</sub>


















































































【0027】

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
17									CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
18									CH <sub>3</sub>	H	H	H
19									OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>
20									OCH <sub>3</sub>	H	H	H
21									SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>
22									SCH <sub>3</sub>	H	H	H
23									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
24									SCH <sub>3</sub>	H	SCH <sub>3</sub>	H

【0028】



[illegible]

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
41												
42		H	H	H								
43			H	H								
44		H		H								
45		H	H									
46	CH <sub>3</sub>	CH <sub>3</sub>										
47	CH <sub>3</sub>	CH <sub>3</sub>										
48	OCH <sub>3</sub>	OCH <sub>3</sub>										

【0031】


















































































化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
48												
50										H	H	H
51											H	H
52										H		H
53										H	H	
54									SCH <sub>3</sub>	SCH <sub>3</sub>		
55									CH <sub>3</sub>	CH <sub>3</sub>		
56									OCH <sub>3</sub>	OCH <sub>3</sub>		

【0032】

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
57												
58										H	H	H
59											H	H
60										H		H
61										H	H	
62									SCH <sub>3</sub>	SCH <sub>3</sub>		
63									CH <sub>3</sub>	CH <sub>3</sub>		
64									OCH <sub>3</sub>	OCH <sub>3</sub>		

【0033】

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
73												
74										H	H	H
75											H	H
76										H		H
77										H	H	
78									CH <sub>3</sub>	CH <sub>3</sub>		
79									OCH <sub>3</sub>	OCH <sub>3</sub>		
80									SCH <sub>3</sub>	SCH <sub>3</sub>		

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
81												
82		H	H	H								
83			H	H								
84		H		H								
85		H	H									
86	CH <sub>3</sub>	CH <sub>3</sub>										
87	OCH <sub>3</sub>	OCH <sub>3</sub>										
88	SCH <sub>3</sub>	SCH <sub>3</sub>										

【0036】

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
89									H	H	H	H
90									H	H	H	H
91									H	H	H	H
92									H	H	H	H
93									H	H	H	H
94											H	H

【0037】

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
95			CH <sub>3</sub>	CH <sub>3</sub>					CH <sub>3</sub>	CH <sub>3</sub>		
96	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
97	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H								
98	CH <sub>3</sub>	CH <sub>3</sub>	H	H					H	H		
99	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
100			CH <sub>3</sub>	CH <sub>3</sub>								

【0038】

17

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
101									H	H	H	H
102									H	H	H	H
103									CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
104									CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
105									CH <sub>3</sub>	CH <sub>3</sub>	H	H
108									CH <sub>3</sub>	CH <sub>3</sub>	H	H

【0039】

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
107									SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>
108									SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>
109									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
110									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
111									OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>
112									OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>

21

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
113									OCH <sub>3</sub>	OCH <sub>3</sub>	H	H
114									OCH <sub>3</sub>	OCH <sub>3</sub>	H	H
115									SCH <sub>3</sub>	SCH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
116									SCH <sub>3</sub>	SCH <sub>3</sub>	OCH <sub>3</sub>	OCH <sub>3</sub>
117									OCH <sub>3</sub>	OCH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
118									OCH <sub>3</sub>	OCH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>

【0041】






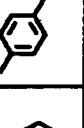




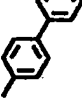
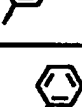














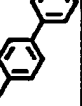















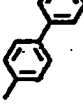
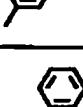




23

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
119												
120												
121												
122												
123												
124												

【0042】





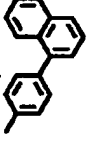
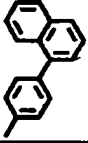




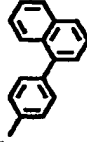
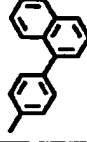








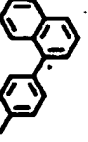
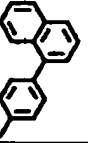




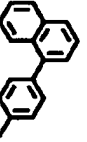
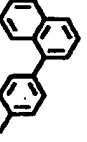








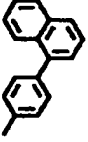
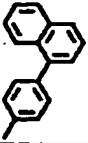




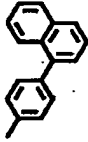
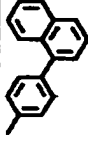






25

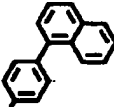

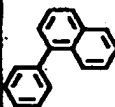

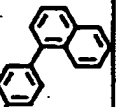

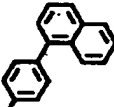

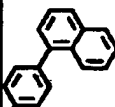

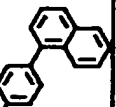

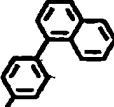

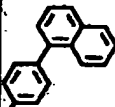

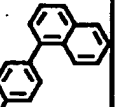

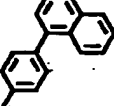

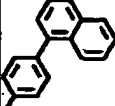

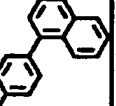


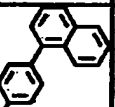

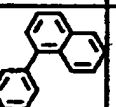

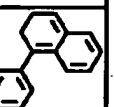

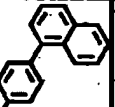

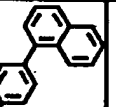



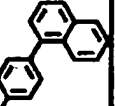

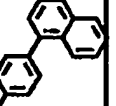

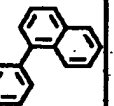

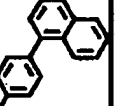

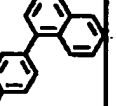

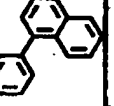
化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
125	C <sub>2</sub> H <sub>5</sub>	C <sub>2</sub> H <sub>5</sub>	H	H								
126	C <sub>2</sub> H <sub>5</sub>	C <sub>2</sub> H <sub>5</sub>	H	H								
127	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
128	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>								
129	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
130	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								

【0043】

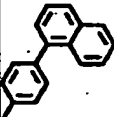

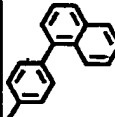

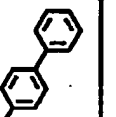

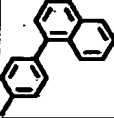

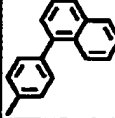

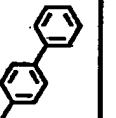

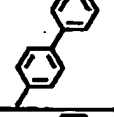

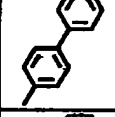

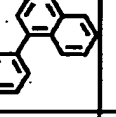

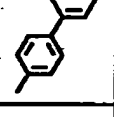

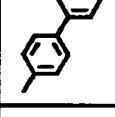

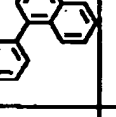


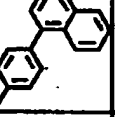

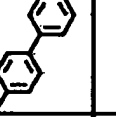

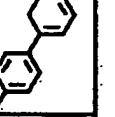

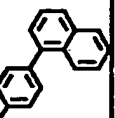

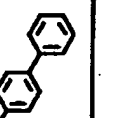

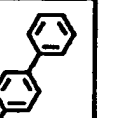

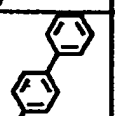

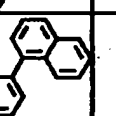

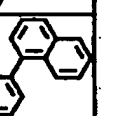

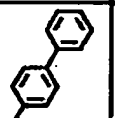

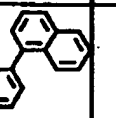

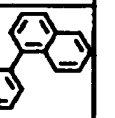
27

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
131	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
132	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
133	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
134	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>	SCH <sub>3</sub>								
135	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
136	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								























































【0044】

$R^{12}$	H	H	H	CH <sub>3</sub>	H	H
$R^{11}$	H	H	H	CH <sub>3</sub>	H	H
$R^{10}$	H	H	CH <sub>3</sub>	OCH <sub>3</sub>	SCH <sub>3</sub>	OCH <sub>3</sub>
$R^9$	H	H	CH <sub>3</sub>	OCH <sub>3</sub>	SCH <sub>3</sub>	OCH <sub>3</sub>
$R^8$						
$R^7$						
$R^6$						
$R^5$						
$R^4$						
$R^3$						
$R^2$						
$R^1$						
化合物	137	138	139	140	141	142

31

















































$R^{12}$	H	H	H	CH <sub>3</sub>	H	H
$R^{11}$	H	H	H	CH <sub>3</sub>	H	H
$R^{10}$	H	H	CH <sub>3</sub>	OCH <sub>3</sub>	SCH <sub>3</sub>	OCH <sub>3</sub>
$R^9$	H	H	CH <sub>3</sub>	OCH <sub>3</sub>	SCH <sub>3</sub>	OCH <sub>3</sub>
$R^8$						
$R^7$						
$R^6$						
$R^5$						
$R^4$						
$R^3$						
$R^2$						
$R^1$						
化合物	143	144	145	146	147	148





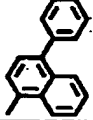
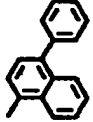
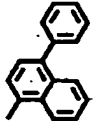
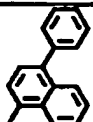
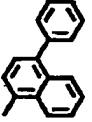
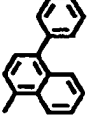
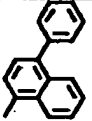
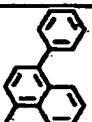








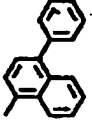
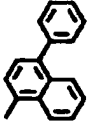
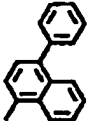
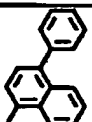
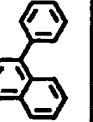
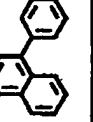
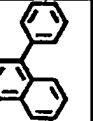
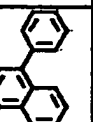








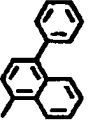
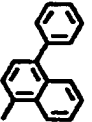
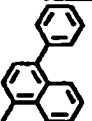
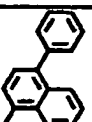
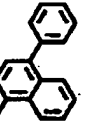
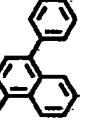
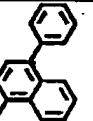
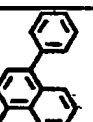
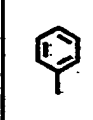



【0046】

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
149	H	H	CH <sub>3</sub>	CH <sub>3</sub>									
150	H	H	CH <sub>3</sub>	CH <sub>3</sub>									
151	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>									
152	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H									
153	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>									
154	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H									





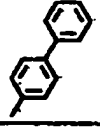
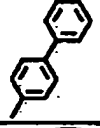
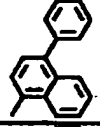
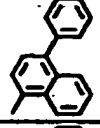
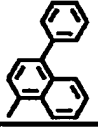
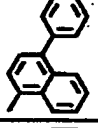
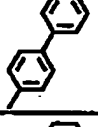
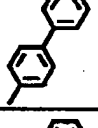
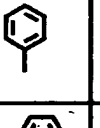

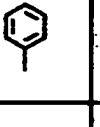





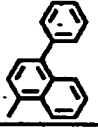
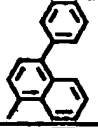
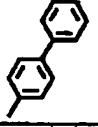
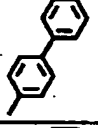
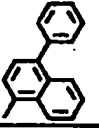
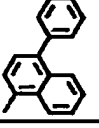
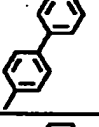
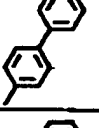


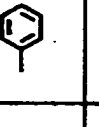





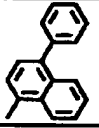
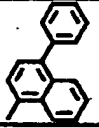
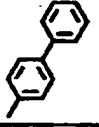
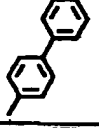
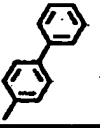
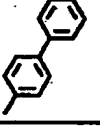
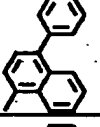
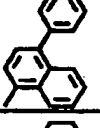




	$\text{CH}_3$	$\text{CH}_3$	$\text{OCH}_3$	H	$\text{SCH}_3$	H
	$\text{CH}_3$	$\text{CH}_3$	$\text{OCH}_3$	H	$\text{SCH}_3$	H
	H	H	H	$\text{OCH}_3$	H	$\text{SCH}_3$
	H	H	H	$\text{OCH}_3$	H	$\text{SCH}_3$
化合物	156	158	157	158	158	160



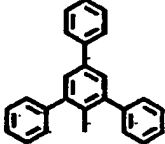
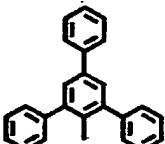










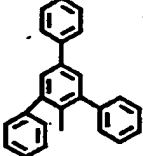
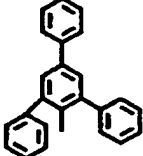


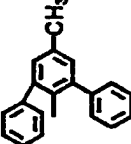
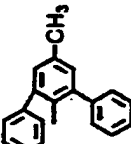










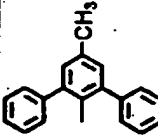
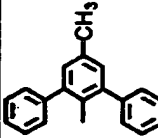


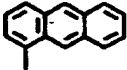
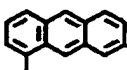




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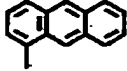
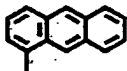




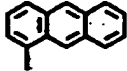


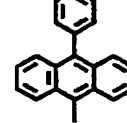
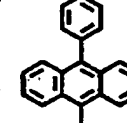







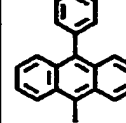
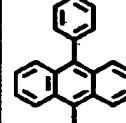



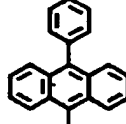



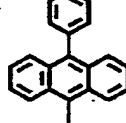
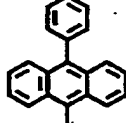



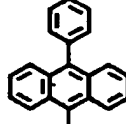



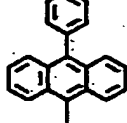
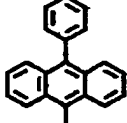





化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
181	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
182	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
183	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								
184	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H								
185	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
186	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								



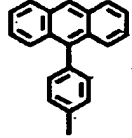
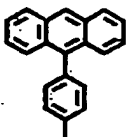


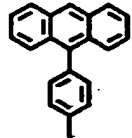
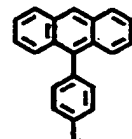


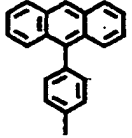
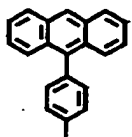






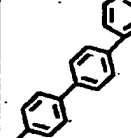
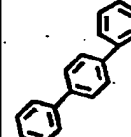






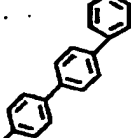
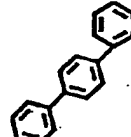


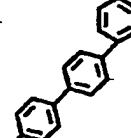
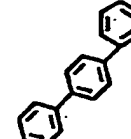


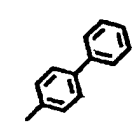
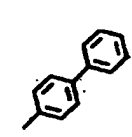


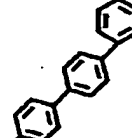
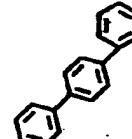
化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
167	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
168	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
169	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								
170	OCH <sub>3</sub>	OCH <sub>3</sub>	H	H								
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172	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								

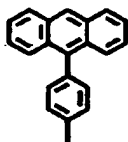
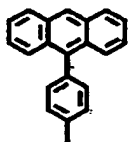
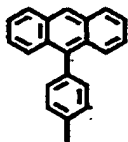
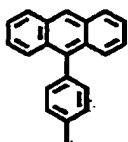





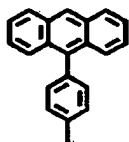
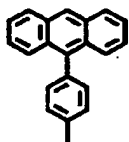







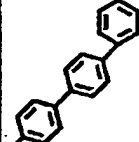
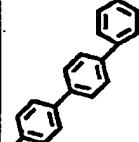







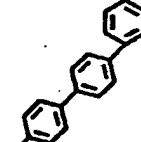
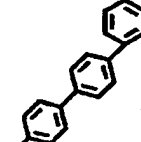
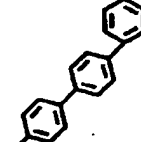
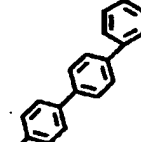
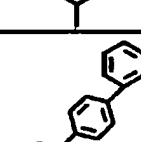
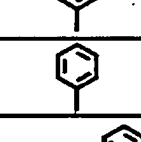
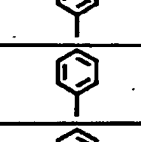
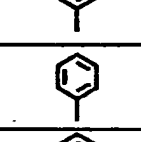
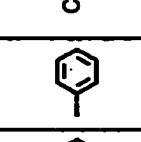
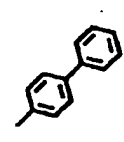
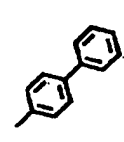
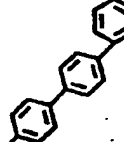
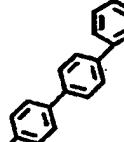
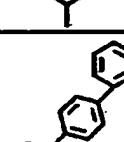
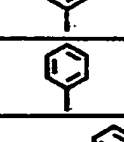
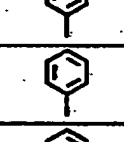
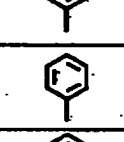
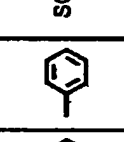


化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
173	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
174	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
175	H	H	OCH <sub>3</sub>	OCH <sub>3</sub>								
176	OCH <sub>3</sub> OCH <sub>3</sub>	OCH <sub>3</sub> OCH <sub>3</sub>	H	H								
177	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
178	SCH <sub>3</sub> SCH <sub>3</sub>	SCH <sub>3</sub> SCH <sub>3</sub>	H	H								

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
179	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
180	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
181	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
182	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
183	CH <sub>3</sub>	CH <sub>3</sub>	H	H								

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
184	CH <sub>3</sub>	CH <sub>3</sub>	H	H									
185	H	H	CH <sub>3</sub>	CH <sub>3</sub>									
186	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H									
187	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>									
188	CH <sub>3</sub>	CH <sub>3</sub>	H	H									





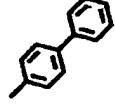
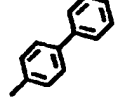


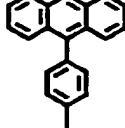
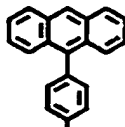




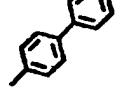
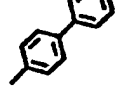








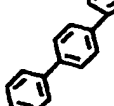
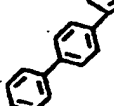








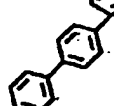
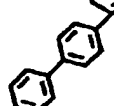


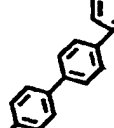
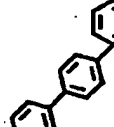




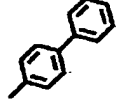
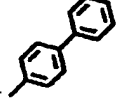


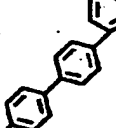
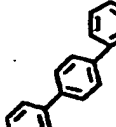
化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
189	CH <sub>3</sub>	CH <sub>3</sub>	H	H								
190	H	H	CH <sub>3</sub>	CH <sub>3</sub>								
191	SCH <sub>3</sub>	SCH <sub>3</sub>	H	H								
192	H	H	SCH <sub>3</sub>	SCH <sub>3</sub>								
193	CH <sub>3</sub>	CH <sub>3</sub>	H	H								

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
194										CH <sub>3</sub>	H	H
195										SCH <sub>3</sub>	H	H
196										H	OCH <sub>3</sub>	OCH <sub>3</sub>
197										CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
198										SCH <sub>3</sub>	H	H

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
199										CH <sub>3</sub>	H	H
200										SCH <sub>3</sub>	H	H
201										H	OCH <sub>3</sub>	OCH <sub>3</sub>
202										CH <sub>3</sub>	CH <sub>3</sub>	CH <sub>3</sub>
203										SCH <sub>3</sub>	H	H



















































R <sup>12</sup>	I	I	I	I	I
R <sup>11</sup>	I	I	I	I	I
R <sup>10</sup>					
R <sup>9</sup>					
R <sup>8</sup>					
R <sup>7</sup>					
R <sup>6</sup>					
R <sup>5</sup>					
R <sup>4</sup>					
R <sup>3</sup>					
R <sup>2</sup>					
R <sup>1</sup>					
化合物	204	205	206	207	208

















































55

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
209	CH <sub>3</sub>	CH <sub>3</sub>										
210	H	H										
211	SCH <sub>3</sub>	SCH <sub>3</sub>										
212	H	H										
213	CH <sub>3</sub>	CH <sub>3</sub>										



























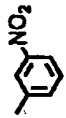







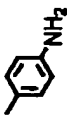





【0058】


























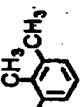
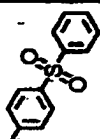



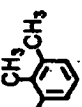
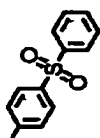
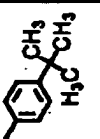
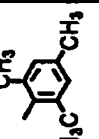
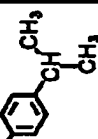


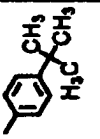
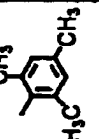
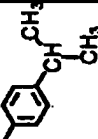


化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
214	CH <sub>3</sub>	CH <sub>3</sub>										
215	H	H										
216	H	H										
217	H	H										
218	CH <sub>3</sub>	CH <sub>3</sub>										

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
219	H	H	CN	CN								
220	H	H	Br	Br								
221	H	H	NO <sub>2</sub>	NO <sub>2</sub>								
222	CN	CN	H	H								
223	H	H	Br	Br								
224	Cl	Cl	H	H								

61

化合物	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>	R <sup>4</sup>	R <sup>5</sup>	R <sup>6</sup>	R <sup>7</sup>	R <sup>8</sup>	R <sup>9</sup>	R <sup>10</sup>	R <sup>11</sup>	R <sup>12</sup>
225									H	H	H	H
226									SCH <sub>3</sub>	SCH <sub>3</sub>	H	H
227									CH <sub>3</sub>	H	CH <sub>3</sub>	CH <sub>3</sub>
228									CH <sub>3</sub>	CH <sub>3</sub>	H	H
229									H	H	H	H

【0061】

R <sup>12</sup>	H	H	CH <sub>3</sub>	H	H
R <sup>11</sup>	H	H	CH <sub>3</sub>	H	H
R <sup>10</sup>	H	SCH <sub>3</sub>	H	CH <sub>3</sub>	H
R <sup>9</sup>	H	SCH <sub>3</sub>	H	CH <sub>3</sub>	H
R <sup>8</sup>					
R <sup>7</sup>					
R <sup>6</sup>					
R <sup>5</sup>					
R <sup>4</sup>					
R <sup>3</sup>					
R <sup>2</sup>					
R <sup>1</sup>					
化合物	230	231	232	234	235

【0062】有機EL素子は、陽極と陰極間に一層もしくは多層の有機化合物薄膜を形成した素子である。一層型の場合、陽極と陰極との間に発光層を設けている。発光層は、発光材料を含有し、それに加えて陽極から注入した正孔もしくは陰極から注入した電子を発光材料まで輸送させるために正孔注入材料、正孔輸送材料もしくは電子注入材料、電子輸送材料を含有しても良い。電子注入材料とは陰極から電子を注入されうる能力を持つ材料\* 50

\*であり、電子輸送材料とは注入された電子を発光層へ輸送する能力を持つ材料である。正孔注入材料とは、陽極から正孔を注入されうる能力を持つ材料であり、正孔輸送材料とは、注入された正孔を発光層へ輸送する能力を持つ材料である。

【0063】多層型としては、陽極/正孔注入帯域/発光層/陰極、陽極/発光層/電子注入帯域/陰極、陽極/正孔注入帯域/発光層/電子注入帯域/陰極の多層構

成で積層した有機EL素子がある。

【0064】本発明の一般式〔1〕で示される化合物は、固体状態において強い蛍光を持つ化合物であり、電界発光性に優れているので、発光材料として発光層内で使用することができる。また、一般式〔1〕の化合物は、発光層内においてドーピング材料として発光層中に最適の割合でドーピングすることにより、高い発光効率および発光波長の最適な選択が可能である。更に、一般式〔1〕の化合物は、正孔もしくは電子等のキャリアを輸送することが出来るが、正孔輸送性がより優れているので、正孔注入層として使用することが出来る。正孔注入層が二層以上で構成される場合、いずれの正孔注入層にも使用することが出来る。

【0065】発光層のホスト材料に、ドーピング材料（ゲスト材料）として一般式〔1〕の化合物を使用して、発光輝度が高い有機EL素子を得ることもできる。一般式〔1〕の化合物は、発光層内において、ホスト材料に対して0.001重量%～50重量%の範囲で含有されていることが望ましく、更には0.01重量%～10重量%の範囲が効果的である。

【0066】一般式〔1〕の化合物と併せて使用できるホスト材料としては、キノリン金属錯体、オキサジアゾール、ベンゾチアゾール金属錯体、ベンゾオキサゾール金属錯体、ベンゾイミダゾール金属錯体、トリアゾール、イミダゾール、オキサゾール、オキサジアゾール、スチルベン、ブタジエン、ベンジジン型トリフェニルアミン、スチルルアミン型トリフェニルアミン、ジアミン型トリフェニルアミンフルオレノン、ジアミノアントラセン型トリフェニルアミン、ジアミノフェナントレン型トリフェニルアミン、アントラキノジメタン、ジフェノキノン、チアジアゾール、テトラゾール、ペリレンテトラカルボン酸、フレオレニリデンメタン、アントラキノジメタン、トリフェニレン、アントロン等とそれらの誘導体、ポリビニルカルバゾール、ポリシラン等の導電性高分子の高分子材料等がある。

【0067】更に、一般式〔1〕の化合物をホスト材料として、他のドーピング材料を使用して発光色を変化させることも可能である。一般式〔1〕と共に使用されるドーピング材料としては、アントラセン、ナフタレン、フェナントレン、ピレン、テトラセン、コロネン、クリセン、フルオレセイン、ペリレン、フタロペリレン、ナフタロペリレン、ペリノン、フタロペリノン、ナフタロペリノン、ジフェニルブタジエン、テトラフェニルブタジエン、クマリン、オキサジアゾール、アルダジン、ビスベンゾキサゾリン、ビススチリル、ピラジン、シクロペンタジエン、キノリン金属錯体、アミノキノリン金属錯体、イミン、ジフェニルエチレン、ビニルアントラセン、ジアミノカルバゾール、ピラン、チオピラン、ポリメチン、メロシアニン、イミダゾールキレート化オキシノイド化合物、キナクリドン、ルブレン等およびそれら

の誘導体があるが、これらに限定されるものではない。

【0068】発光層には、発光材料およびドーピング材料に加えて、必要があれば正孔注入材料や電子注入材料を使用することもできる。

【0069】有機EL素子は、多層構造にすることによりクエンチングによる輝度や寿命の低下を防ぐことができる。また、必要があれば、発光材料、ドーピング材料、キャリア注入を行う正孔注入材料や電子注入材料を二種類以上組み合わせて使用することも出来る。更に、正孔注入層、発光層および電子注入層はそれぞれ二層以上の層構成により形成されていてもよく、正孔もしくは電子が効率よく電極から注入され、効率よく層中で輸送され得る素子構造が選択される。

【0070】有機EL素子の陽極に使用される導電性材料としては、4eVより大きな仕事関数を持つものが適しており、炭素、アルミニウム、バナジウム、鉄、コバルト、ニッケル、タングステン、銀、金、白金、パラジウム等およびそれらの合金、ITO基板、NESA基板と称される酸化スズ、酸化インジウム等の酸化金属、更にはポリチオフェンやポリピロール等の有機導電性樹脂が用いられる。

【0071】陰極に使用される導電性材料としては、4eVより小さな仕事関数を持つものが適しており、マグネシウム、カルシウム、錫、鉛、チタニウム、イットリウム、リチウム、ルテニウム、マンガン等およびそれらの合金が用いられる。合金としては、マグネシウム／銀、マグネシウム／インジウム、リチウム／アルミニウム等が代表例として挙げられるが、これらに限定されるものではない。合金の比率は、加熱の温度、雰囲気、真空度により制御され適切な比率が選択される。陽極および陰極は、必要があれば二層以上の層構成により形成されていても良い。

【0072】有機EL素子では、効率良く発光させるために、用いられる陽極、陰極のうち少なくとも一方は素子の発光波長領域において充分透明であることが望ましい。また、基板もまた透明であることが望ましい。透明電極は、上記の導電性材料を使用して、蒸着やスパッタリング等の方法で所定の透光性を確保するように設定する。発光面の電極は、光透過率を10%以上にするのが望ましい。

【0073】基板は、機械的、熱的強度を有し、透明であれば限定されるものではないが、例示すると、ガラス基板、ポリエチレン板、ポリエーテルサルフォン板、ポリプロピレン板等の透明性樹脂が挙げられる。

【0074】本発明に係る有機EL素子の各層の形成は、真空蒸着、スパッタリング等の乾式成膜法やスピンコーティング、ディッピング等の湿式成膜法の何れの方法でも適用することができる。膜厚は特に限定されるものではないが、各層は適切な膜厚に設定する必要がある。膜が厚すぎると、一定の光出力を得るために大きな

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印加電圧が必要になり効率が悪くなり、膜が薄すぎるとピンホール等が発生し、電圧を印加しても十分な発光輝度が得られない。通常用いられる膜厚としては5 nmから10  $\mu$ mの範囲が適しているが、10 nmから0.2  $\mu$ mの範囲が更に好ましい。

【0075】湿式成膜法の場合には、各層を形成する材料をクロロホルム、テトラヒドロフラン、ジオキサン等の適切な溶媒に溶解あるいは分散して有機化合物薄膜を形成する。

【0076】乾式あるいは湿式成膜法から調製される何れ10の薄膜においても、成膜性向上、ピンホール防止等の目的の為に適切な樹脂あるいは添加剤を使用してもよい。この様な樹脂としては、ポリスチレン、ポリカーボネート、ポリアリレート、ポリエステル、ポリアミド、ポリウレタン、ポリスルホン、ポリメチルメタクリレート、ポリメチルアクリレート、セルロース等の絶縁性樹脂、ポリ-N-ビニルカルバゾール、ポリシラン等の光導電性樹脂、ポリチオフェン、ポリピロール等の導電性樹脂を挙げることができる。また、添加剤としては、酸化防止剤、紫外線吸収剤、可塑剤等を挙げることが20

【0077】正孔注入材料としては、正孔を注入する能力を持ち、発光層または発光材料に対して優れた正孔注入効果を有し、発光層で生成した励起子の電子注入層または電子注入材料への移動を防止し、かつ薄膜形成能の優れた化合物が挙げられる。具体的には、フクロシアニン系化合物、ナフトロシアニン系化合物、ボルフィリン系化合物、オキサジアゾール、トリアゾール、イミダゾール、イミダゾロン、イミダゾールチオン、ピラゾリン、ピラゾロン、テトラヒドロイミダゾール、オキサゾール、オキサジアゾール、ヒドラゾン、アシルヒドラゾン、ポリアリールアルカン、スチルベン、プタジエン、ベンジジン型トリフェニルアミン、スチリルアミン型トリフェニルアミン、ジアミン型トリフェニルアミン等と、それらの誘導体、およびポリビニルカルバゾール、ポリシラン、導電性高分子等の高分子材料等があるが、これらに限定されるものではない。

【0078】電子注入材料としては、電子を注入する能力を持ち、発光層または発光材料に対して優れた電子注入効果を有し、発光層で生成した励起子の正孔注入層または正孔注入材料への移動を防止し、かつ薄膜形成能の優れた化合物が挙げられる。例としては、キノリン金属錯体、オキサジアゾール、ベンゾチアゾール金属錯体、ベンゾオキサゾール金属錯体、ベンゾイミダゾール金属錯体、フルオレノン、アントラキノジメタン、ジフェノキノン、チオピランジオキソド、オキサジアゾール、チアジアゾール、テトラゾール、ペリレンテトラカルボン酸、フレオレニリデンメタン、アントラキノジメタン、アントロン等とそれらの誘導体等が挙げられるが、これらに限定されるものではない。また、正孔注入材料に電

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子受容物質を、電子注入材料に電子供与性物質を添加することによる増感も可能である。

【0079】本発明により得られた有機EL素子の、温度、湿度、雰囲気等に対する安定性の向上の為に、素子の表面に保護層を設けたり、シリコンオイル等を封入して素子全体を保護することも可能である。

【0080】

【実施例】以下に本発明の化合物を用いた実施例を示す。本例では、混合比は全て重量比である。また電極面積2 mm $\times$ 2 mmの有機EL素子の特性を測定した。

【0081】実施例1

洗浄したITO電極付きガラス板上に、発光材料として表1の化合物(19)、2,5-ビス(1-ナフチル)-1,3,4-オキサジアゾール、ポリカーボネート樹脂(帝人化成:バンライトK-1300)を1:2:10の重量比でテトラヒドロフランに溶解させ、スピンコーティング法により膜厚100 nmの発光層を得た。その上に、マグネシウムと銀を10:1(重量比)で混合した合金で膜厚150 nmの電極を形成して有機EL素子を得た。この素子の発光特性は、直流電圧5 Vでの発光輝度50 (cd/m<sup>2</sup>)、最大発光輝度580 (cd/m<sup>2</sup>)、発光効率0.62 (lm/W)の発光が得られた。

【0082】実施例2

洗浄したITO電極付きガラス板上に、N,N'-(3-メチルフェニル)-N,N'-ジフェニル-1,1'-ビフェニル-4,4'-ジアミン(TPD)を真空蒸着して膜厚20 nmの正孔注入層を得た。次いで、表1の化合物(2)を蒸着し膜厚40 nmの発光層を作成し、次いでトリス(8-ヒドロキシキノリナート)アルミニウム錯体(Alq3)を蒸着して膜厚30 nmの電子注入層を得た。その上に、マグネシウムと銀を10:1(重量比)で混合した合金で膜厚100 nmの電極を形成して有機EL素子を得た。各層は10<sup>-4</sup> Paの真空中で、基板温度室温の条件下で蒸着した。この素子は直流電圧5 Vでの発光輝度580 (cd/m<sup>2</sup>)、最大発光輝度13600 (cd/m<sup>2</sup>)、発光効率1.6 (lm/W)の発光が得られた。

【0083】実施例3

40 洗浄したITO電極付きガラス板上に、表1の化合物(31)を塩化メチレンに溶解させ、スピンコーティング法により膜厚50 nmの正孔注入型発光層を得た。次いで、ビス(2-メチル-8-ヒドロキシキノリナート)(1-ナフトラート)ガリウム錯体を真空蒸着して膜厚40 nmの電子注入層を作成し、その上に、マグネシウムと銀を10:1(重量比)で混合した合金で膜厚100 nmの電極を形成して有機EL素子を得た。電子注入層は10<sup>-4</sup> Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5 Vでの発光輝度320 (cd/m<sup>2</sup>)、最大発光輝度9300 (cd/m<sup>2</sup>)

<sup>2</sup>)、発光効率1.3 (lm/W)の発光が得られた。

#### 【0084】実施例4

洗浄したITO電極付きガラス板上に、表1の化合物(41)を真空蒸着して膜厚50nmの正孔注入型発光層を得た。次いで、ビス(2-メチル-8-ヒドロキシキノリナート)(p-シアノフェノラート)ガリウム錯体を真空蒸着して膜厚30nmの電子注入層を作成し、その上に、マグネシウムと銀を10:1(重量比)で混合した合金で膜厚100nmの電極を形成して有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5Vでの発光輝度390 (cd/m<sup>2</sup>)、最大発光輝度13900 (cd/m<sup>2</sup>)、発光効率1.4 (lm/W)の発光が得られた。

#### 【0085】実施例5~19

洗浄したITO電極付きガラス板上に、4, 4'-ビス\*

【表2】

実施例	表1の発光材料	発光輝度 (cd/m <sup>2</sup> )	最大発光輝度 (cd/m <sup>2</sup> )	最大発光効率 (lm/W)
5	(4)	290	17500	1.4
6	(40)	610	19700	1.8
7	(45)	690	18700	1.5
8	(92)	140	12900	1.7
9	(96)	350	17200	1.3
10	(101)	550	17300	2.0
11	(103)	320	12600	1.5
12	(104)	520	19900	1.6
13	(111)	810	11300	1.4
14	(133)	420	11200	2.0
15	(151)	770	11500	1.3
16	(168)	420	18200	1.7
17	(179)	250	10700	1.0
18	(194)	290	18300	1.8
19	(209)	730	16900	1.3
27	(81)	530	18200	2.1
28	(90)	510	26300	2.3
29	(102)	610	27900	2.6
30	(137)	670	19900	1.9
31	(197)	420	20600	2.5

発光輝度は直流5V印加時の値

#### 【0087】実施例20

洗浄したITO電極付きガラス板上に、4, 4', 4'-トリリス[N-(3-メチルフェニル)-N-フェニルアミノ]トリフェニルアミンを真空蒸着して、膜厚40nmの正孔注入層を得た。次いで、 $\alpha$ -NPDを真空蒸着して、膜厚10nmの第二正孔注入層を得た。さらに、表1の化合物(17)を真空蒸着して、膜厚30nmの発光層を作成し、さらにビス(2-メチル-8-ヒドロキシキノリナート)(フェノラート)ガリウム錯体を真空蒸着して膜厚30nmの電子注入層を作成し、その上に、アルミニウムとリチウムを25:1(重量比)で混合した合金で膜厚150nmの電極を形成して、有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5※50

\*[N-(1-ナフチル)-N-フェニルアミノ]ビフェニル( $\alpha$ -NPD)を真空蒸着して膜厚30nmの正孔注入層を形成した。次いで、発光材料として表2に示した材料を真空蒸着して膜厚30nmの発光層を得た。次いで、ビス(2-メチル-8-ヒドロキシキノリナート)(フェノラート)ガリウム錯体を真空蒸着して膜厚30nmの電子注入層を作成し、その上に、マグネシウムと銀を10:1(重量比)で混合した合金で膜厚100nmの電極を形成して有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子の発光特性を表2に示す。ここでの発光輝度は、直流電圧5V印可時の輝度である。本実施例の有機EL素子は、全て最高輝度10000 (cd/m<sup>2</sup>)以上の高輝度特性を有した。

#### 【0086】表2

【表2】

※Vでの発光輝度840 (cd/m<sup>2</sup>)、最大発光輝度13200 (cd/m<sup>2</sup>)、発光効率1.9 (lm/W)の発光が得られた。

#### 【0088】実施例21

洗浄したITO電極付きガラス板上に、4, 4'-ビス[N-(1-ナフチル)-N-フェニルアミノ]ビフェニル( $\alpha$ -NPD)を真空蒸着して膜厚20nmの正孔注入層を得た。次いで、表1の化合物(26)を真空蒸着し膜厚40nmの発光層を作成し、次いでトリリス(8-ヒドロキシキノリナート)アルミニウム錯体(Alq3)を蒸着して膜厚30nmの電子注入層を得た。その上にまず、フッ化リチウム(LiF)を0.5nm、さらにアルミニウム(Al)を200nm真空蒸着によって電極を形成して有機EL素子を得た。各層は $10^{-4}$ P

aの真空中で、基板温度室温の条件下で蒸着した。この素子は直流電圧5Vでの発光輝度300 (cd/m<sup>2</sup>)、最大発光輝度17400 (cd/m<sup>2</sup>)、発光効率1.9 (lm/W)の発光が得られた。

#### 【0089】実施例22

ITO電極と表1の化合物(201)の間に、銅フタロシアニンの膜厚5nmの正孔注入層を設ける以外は、実施例3と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vで発光輝度580 (cd/m<sup>2</sup>)、最大発光輝度16200 (cd/m<sup>2</sup>)、発光効率1.7 (lm/W)の発光が得られた。

#### 【0090】実施例23

4, 4', 4"-トリス[N-(3-メチルフェニル)-N-フェニルアミノ]トリフェニルアミンの代わりに無金属フタロシアニンの膜厚20nmの正孔注入層を設ける以外は、実施例20と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vでの発光輝度560 (cd/m<sup>2</sup>)、最大発光輝度21100 (cd/m<sup>2</sup>)、発光効率2.1 (lm/W)の発光が得られた。

#### 【0091】実施例24

発光層として、表1の化合物(13)と(15)を2:3の重量比率で蒸着した膜厚30nmの薄膜を設ける以外は、実施例5と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vでの発光輝度900 (cd/m<sup>2</sup>)、最大発光輝度21700 (cd/m<sup>2</sup>)、発光効率2.5 (lm/W)の発光が得られた。

#### 【0092】実施例25

発光層として、表1の化合物(24)とトリス(8-ヒドロキシキノリナート)アルミニウム錯体(Alq3)を1:10の重量比率で蒸着した膜厚30nmの薄膜を設ける以外は、実施例5と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vでの発光輝度360 (cd/m<sup>2</sup>)、最大発光輝度18900 (cd/m<sup>2</sup>)、発光効率2.2 (lm/W)の発光が得られた。

#### 【0093】実施例26

発光層として、表1の化合物(68)と4, 4'-ビス(β, β-ジフェニルビニル)ビフェニルを3:100の重量比率で蒸着した膜厚30nmの薄膜を設ける以外は、実施例5と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vでの発光輝度440 (cd/m<sup>2</sup>)、最大発光輝度23500 (cd/m<sup>2</sup>)、発光効率2.5 (lm/W)の発光が得られた。

#### 【0094】実施例27~31

発光層として、表2に示した化合物と4, 4'-ビス[N-(9-フェナントリル)-N-フェニルアミノ]ビフェニルを1:20の重量比率で蒸着した膜厚30nmの薄膜を設ける以外は、実施例5と同様の方法で有機EL素子を作製した。この素子の発光特性を表2に示す。ここでの発光輝度は、直流電圧5V印可時の輝度で

ある。本実施例の有機EL素子は、全て最高輝度15000 (cd/m<sup>2</sup>)以上の高輝度特性を有した。

#### 【0095】実施例32

発光層として、表1の化合物(227)と4-(ジシアノメチレン)-2-メチル-6-(p-ジメチルアミノスチリル)-4H-ピラン(DCM)を100:3の重量比率で蒸着した膜厚30nmの薄膜を設ける以外は、実施例5と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vでの発光輝度880 (cd/m<sup>2</sup>)、最大発光輝度15000 (cd/m<sup>2</sup>)、発光効率1.8 (lm/W)の発光が得られた。

#### 【0096】実施例33

洗浄したITO電極付きガラス板上に、銅フタロシアニンを真空蒸着して、膜厚50nmの正孔注入層を得た。次いで、α-NPDを真空蒸着して、膜厚30nmの第二正孔注入層を得た。さらに、表1の化合物(73)を真空蒸着して、膜厚30nmの発光層を作成し、さらにトリス(8-ヒドロキシキノリナート)アルミニウム錯体を真空蒸着して膜厚30nmの電子注入層を作成し、その上に、アルミニウムとリチウムを25:1(重量比)で混合した合金で膜厚150nmの電極を形成して、有機EL素子を得た。各層は10<sup>-4</sup>Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5Vでの発光輝度740 (cd/m<sup>2</sup>)、最大発光輝度18700 (cd/m<sup>2</sup>)、発光効率2.4 (lm/W)の発光が得られた。また、この素子を3 (mA/cm<sup>2</sup>)で連続発光させたところ、3000時間以上安定した発光を観測できた。

#### 【0097】実施例34

洗浄したITO電極付きガラス板上に、表1の化合物(19)を塩化メチレンに溶解させ、スピンコーティング法により膜厚50nmの正孔注入層を得た。次いでビス(2-メチル-8-ヒドロキシキノリナート)(フェノラート)ガリウム錯体を蒸着して膜厚70nmの電子注入型発光層を得た。マグネシウムと銀を10:1(重量比)で混合した合金で膜厚100nmの電極を形成して有機EL素子を得た。各層は10<sup>-4</sup>Paの真空中で、基板温度室温の条件下で蒸着した。この素子は直流電圧5Vでの発光輝度940 (cd/m<sup>2</sup>)、最大発光輝度15900 (cd/m<sup>2</sup>)、発光効率1.7 (lm/W)の発光が得られた。

#### 【0098】実施例35

洗浄したITO電極付きガラス板上に、銅フタロシアニンを真空蒸着して、膜厚50nmの正孔注入層を得た。次いで、表1の化合物(49)を真空蒸着して、膜厚30nmの第二正孔注入層を得た。さらに、N, N, N', N'-テトラキス[p-(α, α-ジメチルベンジル)フェニル]-9, 10-アントラセンジアミンを真空蒸着して、膜厚30nmの発光層を作成し、さらにトリス(8-ヒドロキシキノリナート)アルミニウム錯



体を真空蒸着して膜厚30nmの電子注入層を作成し、その上に、アルミニウムとリチウムを25:1(重量比)で混合した合金で膜厚150nmの電極を形成して、有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5Vでの発光輝度670( $\text{cd}/\text{m}^2$ )、最大発光輝度24500( $\text{cd}/\text{m}^2$ )、発光効率2.6( $\text{lm}/\text{W}$ )の発光が得られた。

#### 【0099】実施例36

洗浄したITO電極付きガラス板上に、表1の化合物(57)を真空蒸着して膜厚30nmの正孔注入層を得た。次いで、 $\alpha$ -NPDとルブレンを重量比100:3で共蒸着して膜厚40nmの発光層を作成し、次いでトリス(8-ヒドロキシキノリナート)アルミニウム錯体(Alq3)を蒸着して膜厚30nmの電子注入層を得た。その上にまず、フッ化リチウム(LiF)を0.5nm、さらにアルミニウム(Al)を200nm真空蒸着によって電極を形成して有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子は直流電圧5Vでの発光輝度740( $\text{cd}/\text{m}^2$ )、最大発光輝度16400( $\text{cd}/\text{m}^2$ )、発光効率1.7( $\text{lm}/\text{W}$ )の発光が得られた。

#### 【0100】実施例37

洗浄したITO電極付きガラス板上に、表1の化合物(65)を真空蒸着して、膜厚40nmの正孔注入層を得た。次いで、 $\alpha$ -NPDを真空蒸着して、膜厚10nmの第二正孔注入層を得た。さらに、トリス(8-ヒドロキシキノリナート)アルミニウムと3-(2'-ベンゾチアゾリル)-7-ジエチルアミノクマリン(クマリン6)を重量比100:2で共蒸着して膜厚30nmの発光層を作成し、さらにビス(2-メチル-8-ヒドロキシキノリナート)(フェノラート)ガリウム錯体を真空蒸着して膜厚30nmの電子注入層を作成し、その上に、アルミニウムとリチウムを25:1(重量比)で混合した合金で膜厚150nmの電極を形成して、有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5Vでの発光輝度860( $\text{cd}/\text{m}^2$ )、最大発光輝度28100( $\text{cd}/\text{m}^2$ )、発光効率2.3( $\text{lm}/\text{W}$ )の発光が得られた。

#### 【0101】実施例38

洗浄したITO電極付きガラス板上に、表1の化合物(199)を真空蒸着して、膜厚40nmの正孔注入層を得た。次いで、表1の化合物(49)を真空蒸着して、膜厚10nmの第二正孔注入層を得た。さらに、4,4'-ビス( $\beta$ , $\beta$ -ジフェニルビニル)ビフェニルを真空蒸着して膜厚30nmの発光層を作成し、さらにビス(2-メチル-8-ヒドロキシキノリナート)(フェノラート)ガリウム錯体を真空蒸着して膜厚30nmの電子注入層を作成し、その上に、マグネシウムと

銀を10:1(重量比)で混合した合金で膜厚100nmの電極を形成して有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5Vでの発光輝度380( $\text{cd}/\text{m}^2$ )、最大発光輝度24300( $\text{cd}/\text{m}^2$ )、発光効率2.4( $\text{lm}/\text{W}$ )の発光が得られた。

#### 【0102】実施例39

洗浄したITO電極付きガラス板上に、4,4'-ビス[N-(1-ナフチル)-N-フェニルアミノ]ビフェニル( $\alpha$ -NPD)を真空蒸着して膜厚30nmの正孔注入層を形成した。次いで、トリス(8-ヒドロキシキノリナート)アルミニウムと3-(2'-ベンゾチアゾリル)-7-ジエチルアミノクマリン(クマリン6)を重量比100:2で共蒸着して膜厚30nmの発光層を得た。次いで、表1の化合物(204)を真空蒸着して膜厚30nmの電子注入層を作成し、その上に、アルミニウムとリチウムを25:1(重量比)で混合した合金で膜厚150nmの電極を形成して有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5Vでの発光輝度290( $\text{cd}/\text{m}^2$ )、最大発光輝度19700( $\text{cd}/\text{m}^2$ )、発光効率2.6( $\text{lm}/\text{W}$ )の発光が得られた。

#### 【0103】実施例40

洗浄したITO電極付きガラス板上に、2,3,6,7,10,11-ヘキサメトキシトリフェニレンを真空蒸着して膜厚30nmの正孔注入層を形成した。次いで、N,N,N',N'-テトラ-p-ビフェニル-1,4-ナフタレンジアミンを真空蒸着して膜厚30nmの発光層を得た。次いで、表1の化合物(217)を真空蒸着して膜厚10nmの第二電子注入層を作成し、さらにビス(2-メチル-8-ヒドロキシキノリナート)(フェノラート)ガリウム錯体を真空蒸着して膜厚30nmの第一電子注入層を作成した。その上に、マグネシウムと銀を10:1(重量比)で混合した合金で膜厚100nmの電極を形成して有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5Vでの発光輝度760( $\text{cd}/\text{m}^2$ )、最大発光輝度25400( $\text{cd}/\text{m}^2$ )、発光効率2.9( $\text{lm}/\text{W}$ )の発光が得られた。

#### 【0104】実施例41

洗浄したITO電極付きガラス板上に、4,4'-ビス[N-(9-フェナントリル)-N-フェニルアミノ]ビフェニルを真空蒸着して膜厚30nmの正孔注入層を形成した。次いで、表1の化合物(235)とN,N'-ジメチルキナクリドンを重量比150:1で共蒸着して膜厚40nmの発光層を得た。次いで、表1の化合物(227)を真空蒸着して膜厚30nmの電子注入層を作成した。その上に、マグネシウムと銀を10:1(重

量比)で混合した合金で膜厚100nmの電極を形成して有機EL素子を得た。各層は $10^{-4}$ Paの真空中で、基板温度室温の条件下で蒸着した。この素子は、直流電圧5Vでの発光輝度710( $\text{cd}/\text{m}^2$ )、最大発光輝度35000( $\text{cd}/\text{m}^2$ )、発光効率4.6( $\text{lm}/\text{W}$ )の発光が得られた。

#### 【0105】比較例1

発光層として、5, 6, 11, 12-テトラフェニルナフタセン(ルブレナ)を真空蒸着した膜厚30nmの薄膜を設ける以外は、実施例5と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vでの発光輝度50( $\text{cd}/\text{m}^2$ )、最大発光輝度1880( $\text{cd}/\text{m}^2$ )、発光効率0.3( $\text{lm}/\text{W}$ )の発光であった。

#### 【0106】比較例2

発光層として、5, 6, 11, 12-テトラビフェニル-2, 3, 8, 9-テトラフェニルナフタセンを真空蒸着した膜厚30nmの薄膜を設ける以外は、実施例20と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vでの発光輝度270( $\text{cd}/\text{m}^2$ )、最大発光輝度2400( $\text{cd}/\text{m}^2$ )、発光効率0.25( $\text{lm}/\text{W}$ )の発光であった。

#### 【0107】比較例3

発光層として、5, 6, 11, 12-テトラビフェニルナフタセンと4, 4'-ビス[N-(9-フェナントリル)-N-フェニルアミノ]ビフェニルを1:20の重量比率で蒸着した膜厚30nmの薄膜を設ける以外は、実施例27と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vでの発光輝度340( $\text{cd}/\text{m}^2$ )、最大発光輝度7200( $\text{cd}/\text{m}^2$ )、発光効率0.95( $\text{lm}/\text{W}$ )の発光であった。

#### 【0108】比較例4

発光層として、2, 3, 5, 6, 8, 9, 11, 12-オクタフェニルナフタセンとDCMを100:3の重量比率で蒸着した膜厚30nmの薄膜を設ける以外は、実施例32と同様の方法で有機EL素子を作製した。この素子は、直流電圧5Vでの発光輝度170( $\text{cd}/\text{m}^2$ )、最大発光輝度5400( $\text{cd}/\text{m}^2$ )、発光効率0.85( $\text{lm}/\text{W}$ )の発光であった。

【0109】本実施例で示された有機EL素子は、二層型以上の素子構成において、最大発光輝度10000( $\text{cd}/\text{m}^2$ )以上の発光が得られ、全て高い発光効率を得ることができた。本実施例で示された有機EL素子について、3( $\text{mA}/\text{cm}^2$ )で連続発光させたところ、1000時間以上安定な発光を観測することができた。

【0110】本発明の有機EL素子は発光効率、発光輝度の向上と長寿命化を達成するものであり、併せて使用される発光材料、ドーピング材料、正孔注入材料、電子注入材料、増感剤、樹脂、電極材料等および素子作製方法を限定するものではない。

#### 【0111】

【発明の効果】本発明の有機EL素子用材料に用いて作成した有機EL素子は、従来に比べて高い発光効率で高輝度であり、長い発光寿命を持つ。特に発光材料に使用した場合には、非常に高特性の素子を作成でき、壁掛けテレビ等のフラットパネルディスプレイや平面発光体として好適に使用することができる。故に、複写機やプリンター等の光源、液晶ディスプレイや計器類等の光源、表示板、標識灯等への応用が可能である。長寿命の有機EL素子を得ることができる。